United States Court of Appeals
for the
District of Columbia Circuit

TRANSCRIPT OF RECORD

BRIEF FOR PLAINTIFF-APPELLANT.

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In the
United States Court of Appeals
DISTRICT OF COLUMBIA CIRCUIT

United States Court of Appeals
For the
District of Columbia Circuit
FILED AUG 22 1958
Joseph W. Stewart
CLERK

Appeal No. 14,550

ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY, A NOT-FOR-PROFIT CORPORATION ORGANIZED UNDER THE LAWS OF THE STATE OF ILLINOIS AND HAVING AN OFFICE AND PLACE OF BUSINESS AT CHICAGO, ILLINOIS,

Plaintiff-Appellant,

vs.

ROBERT C. WATSON, COMMISSIONER OF PATENTS OF THE UNITED STATES, HAVING HIS OFFICIAL RESIDENCE IN THE DISTRICT OF COLUMBIA,

Defendant-Appellee.

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STATEMENT OF QUESTIONS PRESENTED.

1. One question is whether the issuance of a relatively broad patent bars the issuance of a later patent which discloses an improvement over the earlier issued patent.

2. Another question is whether an issued patent claim should be examined in the context of the specification from which it arose where the rejection of a claim in a co-pending application of the same inventor is based upon double patenting.

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JURISDICTIONAL STATEMENT.

This proceeding arises under Title 35, Section 145 of the United States Code, against the Commissioner of Patents to authorize the Commissioner to issue a patent to the Plaintiff-Appellant herein as assignee of Marvin Camras, the inventor and applicant.

STATEMENT OF CASE.

The subject matter of this action arises from proceedings held in the United States Patent Office on applications filed in the Patent Office by one Marvin Camras, a distinguished scientist and inventor (Jt. App. pp. 16-17).

On July 25, 1947, Mr. Camras filed an application for Letters Patent in the United States Patent Office, Serial No. 763,486, dealing with a basic improvement in the manufacture of magnetic iron oxides and of magnetic recording tapes made with such oxides. Prior to the Camras invention, the best magnetic recording media available were metallic ribbons or wires. Low coercivity oxide coated tapes of the prior art gave inferior results and had to be operated at excessively high speeds to record high frequency notes with any degree of fidelity. Mr. Camras, in the invention described in Serial No. 763,486, provided a method for treating particles of iron oxide in very finely divided form to impart magnetic properties to such iron oxide adapting the same for use as a superior magnetic recording medium. The particles treated according to the Camras process are mixed with a suitable binder and applied to a flexible base, such as paper or plastic, and used in that form as a high fidelity magnetic sound recording tape. The overwhelming majority of all magnetic recording tapes on the market today are made in accordance with the invention described in Camras Serial No. 763,486 (Jt. App. p. 20).

Basically, the process described in Camras Serial No. 763,486 involves producing a synthetic iron oxide in finely divided particle form and then reducing the oxide, that is, treating the oxide with hydrogen or other gas capable of removing some of the oxygen of the oxide until the desired magnetic properties are achieved. This reducing step may be followed by oxidation, or by additional oxidation and reduction steps to achieve further benefits.

During the pendency of Camras application Serial No. 763,486 Mr. Camras discovered that certain types of iron oxide materials would not respond adequately to the type of treatment described in the then pending application Serial No. 763,486 to produce completely acceptable magnetic sound recording members. In particular, he discovered that particles which have a cubic particle shape, when treated in accordance with the process of his pending patent application would not achieve magnetic properties as desirable for magnetic recording purposes as achieved by particles having an acicular or needle-like shape. The latter type particles are those which are comprehensively disclosed in his application Serial No. 763,486.

Mr. Camras found that this deficiency in magnetic properties between the two types of materials could be overcome by pre-conditioning the cubic type particles prior to subjecting them to the reduction treatment specified in the prior application. The pre-conditioning treatment consisted of subjecting the particles to a temperature between 1000° and 1750° F. This high temperature treatment is a remarkably novel concept because oxides of iron, when subjected to such high temperatures, lose whatever magnetization they may have previously possessed. Yet, when this high temperature pre-treatment is combined with the reduction steps of the prior Camras invention, it enables materials, which would otherwise respond only incompletely or not at all to the prior Camras process steps, to achieve magnetic properties comparable to those of the very active acicular or needle-like particles.

Mr. Camras then proceeded to file a patent application directed to this pre-treatment step. That application, bearing Serial No. 114,677 and a filing date of September 8, 1949, is the one here in suit. It is this application on which a patent has been refused by the United States Patent Office.

During the pendency of Camras Serial No. 114,677, the prior application Serial No. 763,486 issued as United States Letters Patent No. 2,694,656 on November 16, 1954. The United States Patent Office took the position that the claims of the later filed application Serial No. 114,677 were unpatentable because (1) the subject matter of the later invention was already disclosed in the claimed subject matter of the issued patent, or (2) because the claimed subject matter of Serial No. 114,677 was unpatentable to the applicant in view of his issued patent in combination with references taken from the prior art.

After your petitioner had exhausted its remedies in the United States Patent Office, it filed a complaint under Title 35, U. S. A. Section 145 (Jt. App. pp. 1-4). The Commissioner of Patents filed an answer on July 3, 1956 (Jt. App. p. 5). The District Court for the District of Columbia, through Judge McGarraghy in a memorandum decision dated March 31, 1958 (Jt. App. pp. 36-38) dismissed the complaint. Accordingly, judgment was entered dismissing the complaint (Jt. App. p. 38) and this appeal was lodged (Jt. App. p. 42).

STATUTE INVOLVED.

35 U. S. C. A. Section 145. Civil Action to obtain patent.

> "An applicant dissatisfied with the decision of the Board of Appeals may unless appeal has been taken to the United States Court of Customs and Patent Appeals, have remedy by civil action against the Commissioner in the United States District Court for the District of Columbia if commenced within such time after such decision, not less than sixty days, as the Commissioner appoints. The court may adjudge that such applicant is entitled to receive a patent for his invention, as specified in any of his claims involved in the decision of the Board of Appeals, as the facts in the case may appear and such adjudication shall authorize the Commissioner to issue such patent on compliance with the requirements of law. All the expenses of the proceedings shall be paid by the applicant. July 19, 1952, c. 950 Sec. 66 Stat. 803."

STATEMENT OF POINTS.

1. The District Court, and the United States Patent Office, erred in their interpretation of the claims of Camras Patent No. 2,694,656, particularly in view of the specification of that patent.

2. The District Court in the Patent Office erred in combining the claimed subject matter of Camras Patent No. 2,694,656 with otherwise unrelated references from the prior art in rejecting the claims in Camras Patent Application Serial No. 114,677.

SUMMARY OF ARGUMENT.

First, the lower court erred in its interpretation of the patented claims in Patent No. 2,694,656 insofar as it held that such claims suggested a high temperature pre-treatment of iron oxides for use in the manufacture of magnetic sound recording members.

Second, the lower court erred in combining the claimed subject matter of Camras Patent No. 2,694,656 with other references of the prior art since these other references are not directed to manufacture of magnetic iron oxide particles suitable for use in magnetic recording media, and such combination can be made only with a prior knowledge of the applicant's disclosure in the instant application Serial No. 114,677.

Third, the lower court erred in assuming that Mr. Camras was not entitled to an additional patent for his new improvement for the reason that Mr. Camras' basic Patent No. 2,694,656 has claims which dominate the new process.

ARGUMENT.

This proceeding involves the refusal of the United States Patent Office to grant a patent to Marvin Camras in the field of magnetic recording materials intended for use in the manufacture of magnetic recording tapes.

Mr. Camras' inventions have made the field of magnetic recording a practical art. The earliest types of magnetic recording materials were usually in the form of wire or metallic tape. These media, however, had to be run at very high speeds and evidenced poor quality (Jt. App. p. 19).

A superior, commercially practical, magnetic recording tape, consisting of a base of paper or plastic material covered with a very thin coating of a powdered magnetic iron oxide having desired magnetic recording properties, was developed by Mr. Camras. A description of the method for the preparation of this tape will be found in the issued Camras Patent No. 2,694,656 (Jt. App. pp. 93-99). In this patent, Mr. Camras disclosed that synthetic iron oxide materials of a very small particle size, and preferably acicular or needle-like in configuration, could be rendered suitable for use as magnetic recording media by treating the particles in a reducing atmosphere, that is, an atmosphere of hydrogen gas or the like. With this process, Mr. Camras was able to achieve magnetic properties approximately three times as high as possible with earlier known materials (Jt. App. p. 19). At the present time, about 95% of the magnetic recording tapes sold in this country are made in accordance with the process described in this patent (Jt. App. p. 20).

While the process described in the issued Camras patent will enhance the magnetic recording properties of various iron oxides, it is particularly effective upon those having an acicular or a needle-like crystalline form. Iron oxides of such crystalline form are produced by providing a solution of an iron salt, adding an alkali to precipitate an iron oxide and then using the finely divided precipitate so formed as the nuclei for growing in solution acicular crystals. The chemical composition of the iron oxide, as produced, contains two atoms of iron, three atoms of oxygen, and a molecule of water intimately attached to the molecule of iron oxide. The chemical structure of this material may thus be written $Fe_2O_3 \cdot H_2O$. This material is inherently non-magnetic as produced (Jt. App. pp. 20-21).

Mr. Camras subsequently discovered that some oxides of iron do not respond to the reduction treatment specified in his Patent No. 2,694,656 nearly so well as the preferred acicular particles. In particular, he found that a cubic type of oxide, that is, one whose particles are generally equant in contrast to the elongated needle-type shape, is not sufficiently benefited by the reduction treatment to be competitively useful as commercial magnetic recording materials of the Camras patent. The cubic type of oxide, in contrast to the needle shape oxide contains three atoms of iron and four atoms of oxygen. It contains no water of hydration such as does the needle-shaped oxide. Its chemical formula is Fe_3O_4. The cubic type particles differ from the needle shaped particles additionally in the fact that the cubic material, as prepared, is slightly magnetic (Jt. App. p. 22).

After considerable experimentation, Mr. Camras found that the cubic type particles could be made to respond to the reduction treatment and yield a commercially acceptable magnetic iron oxide having magnetic properties comparable to the needle shaped crystals, by giving the cubic particles a preliminary high temperature treatment at temperatures ranging from about 1000° to 1750° F. The cubic material, when subjected to such high temperatures loses whatever magnetization it has. Nevertheless, this high temperature treatment conditions the material for subsequent reduction in hydrogen gas to produce a material having greatly enhanced magnetic recording properties.

In a sense, the overall process represents an anomaly. Mr. Camras starts with a material which is already somewhat magnetic and rather than going through steps such as reduction which might improve and enlarge upon the magnetic properties already present, the magnetic properties are destroyed and then reappear to a much greater degree when subsequently treated with hydrogen gas. These properties are considerably better than those which can be obtained in the absence of the high temperature treatment. The high temperature treatment step forms an essential step in the process described in the pending Camras Patent Application Serial No. 114,677 involved in this suit. The specification of the Camras application (Jt. App. pp. 45-54) demonstrates the remarkably improved results which are obtained through the use of this high temperature treatment.

To summarize, the invention described in Camras Application Serial No. 114,677 is an improvement upon that described in the issued Patent No. 2,694,656 in that it makes available a new class of starting material for use in the patented process.

Camras Application Serial No. 144,677 and the application which issued as Camras Patent No. 2,694,656 were copending in the Patent Office. Consequently, only the claims of the issued patent are available as references against the claims presented in the Application Serial No. 114,677. We must look, then, to the disclosure in the claims of the issued patent to determine what these claims taught the art.

The Patent Office relied heavily upon the disclosure of claim 13 of the issued patent which reads as follows:

> "13. The method of making magnetic iron oxide which comprises providing a synthetic non-magnetic ferric oxide in crystalline form and of a particle size not over 6 microns in its greatest dimension and reducing said non-magnetic oxide at elevated temperature to produce a ferrosoferric oxide having permanent magnet properties including a coercive force of at least 200 oersteds."

The Patent Office took the position that the phrase "providing a synthetic non-magnetic ferric oxide" discloses to the art the step of intentional demagnetization of an already magnetic iron oxide by a high temperature treatment. We submit that the argument is clearly erroneous both in law and in fact.

To begin with, we do not question that claim 13, recited above, may be broad enough to read upon the new improvement since it is directed essentially to the reduction step, and that step is common to both processes. However, this fact by itself does not raise the question of double patenting. If it did, then no inventor could ever hope to secure an improvement patent once he had issued a broad patent in the field. A noted authority on the subject has stated unequivocally:

> "One of the simplest, clearest, soundest and most essential principles of patent law is that a later invention may be validly patented, although dominated by an earlier patent, whether to the same or to a different inventor. No one will seriously deny the correctness of this statement, in principle."
>
> *Stringham "Double Patenting" Sec. 2831.*

The question is not, as the trial court apparently assumed, whether the claim in the prior patent is sufficiently broad to *dominate* the new improvement but rather whether the claim *discloses* the subject matter of the new invention.

A patent claim is always read in the light of the specification in which it arose. This basic principle of patent law is applied in double patenting situations, too, as indicated in the case of *In re Greenlee* (CCPA 1955) 42 CCPA 926; 222 F. 2d 739; 106 U. S. P. Q. 104.

Applying that principle to the case at bar, the step of "providing a synthetic non-magnetic ferric oxide" as recited in claim 13 must be read in the light of the specific disclosure in the applicant's specification. A reading of the specification of that patent (Jt. App. pp. 94-97) clearly indicates that by the word "providing" was meant the step involved in producing the initial crystals from a solution, or an equivalent of such step. It was so intended by the patentee (Jt. App. pp. 20-21). The "provided" oxide is non-magnetic and necessarily so if prepared as described in the patent. Therefore the phrase "providing a synthetic non-magnetic ferric oxide" as employed in claim 13 is merely the statement of an *existing initial condition* and not an *instruction* to the art that an already magnetic material should be demagnetized. The four corners of the issued patent do not contain any disclosure whatever of a step designed to demagnetize an already feebly magnetic material. This construction of the word "providing" stems only from an attempt of the Patent Office to torture that word into a meaning which it would not otherwise have.

The Patent Office has also relied upon claim 21 of the issued Camras patent as providing a basis for the invention of the high temperature treatment step. Claim 21 of the issued patent reads as follows:

> "21. The method of making permanent magnet

> material, which comprises precipitating crystals of $Fe_2O_3 . H_2O$ from solution of a particle size of less than 6 microns in greatest dimension, heating said crystals in the air to a temperature between 500 and 1550°F. to drive off the water of hydration, reducing said dehydrated crystals at a temperature between 500 and 1000°F. in a gaseous reducing atmosphere to obtain a magnetic material also in crystalline form and of a particle size less than 6 microns and consisting essentially of Fe_3O_4 particles having a coercive force value of at least 200 oersteds and a Bfm/Br (H=1000) of not over 3 to 1."

The Patent Office in making this interpretation overlooks two important considerations. One, claim 21 specifically calls for a precipitating step, which, as is clear from the specification, inherently produces the ferric oxide monohydrate which is the acicular or needle-type form. This material is amenable without any pre-treatment to the reduction treatment to produce acceptable magnetic recording material (Jt. App. pp. 24-25).

Secondly, the claim specifically states that the function of heating at a temperature between 500 and 1500° F. is "to drive off the water of hydration". This further limits the claim to the acicular ferric oxide monohydrate. The starting materials with which the present invention deals are not hydrates so any dehydration treatment would be inapplicable to them (Jt. App. p. 22). Claim 21 of the issued patent, therefore, is directed to the treatment of an entirely different material for an entirely different purpose.

The claims before this court (Jt. App. pp. 2-3) clearly are confined to the present improvement in that they specify the use of a material which, in the absence of the preliminary high temperature treatment, cannot be substantially benefited by the reduction treatment forming the heart of the applicant's prior invention. In other words, the new invention of Mr. Camras adds to the fund of existing knowledge by providing a high temperature treatment in addition to the reduction treatment of the patent and thereby provides the public with a source of magnetic recording material which would be otherwise unavailable.

In this respect, the case at bar is identical to the fact situation presented in the case of *Research Products Co.* v. *Tretolite Co.* (C. A. 9, 1939) 106 F. 2d 530, 535. There the court held that since the later patent expressly excluded from its terms the types of materials covered by the prior patent, no double patenting could exist.

The prior art references cited by the Patent Office (Jt. App. pp. 101-110), need not be considered in detail. These references merely show that it is old to demagnetize iron oxide material by high temperature treatment. The references have nothing to do with magnetic recording (Jt. App. pp. 28-29). They do not suggest a process in which an intentional, high temperature demagnetization treatment is employed as an essential preliminary step to imparting of satisfactory magnetic recording properties to an already magnetic iron oxide by a reduction treatment (Jt. App. pp. 29-30).

The situation in the instant case falls directly in line with the decision in *Hope Basket Co.* v. *Products Advancement Corp.*, (C. A. 6, 1951), 187 F. 2d 1008 where the court stated, on page 1012:

> "However, it is well settled law, as stated in that case, 'where the second patent covers matter described in the prior patent, essentially distinct and separable from the invention covered thereby, and claims made thereunder, its validity may be sustained.' As an example, it is not double patenting when an inventor makes a new improvement on his invention of a patentable character, for which he may obtain a separate patent."

To the same effect, see *Oliver United Filters* v. *Silver* (C. A. 10, 1953) 206 F. 2d 658; Certiorari denied, 346 U. S. 923.

Accordingly, we respectfully submit that the Trial Court and the Patent Office erred in their interpretation of the issued Camras patent and its relation to the pending application; that the decision of the trial court was manifestly contrary to the evidence presented; and that the District Court should have ordered the issuance of the patent under the provisions of Title 35, U. S. C. A. 145.

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BRIEF FOR APPELLEE

United States Court of Appeals
FOR THE DISTRICT OF COLUMBIA CIRCUIT

APPEAL NO. 14,550

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v.

ROBERT C. WATSON, COMMISSIONER OF PATENTS, APPELLEE.

APPEAL FROM THE JUDGMENT OF THE UNITED STATES DISTRICT COURT FOR THE DISTRICT OF COLUMBIA

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STATEMENT OF QUESTIONS PRESENTED

In the opinion of the appellee the questions presented by this appeal are as follows:

1. Whether or not an inventor who has received a patent claiming a process of (a) "providing" a synthetic non-magnetic ferric oxide in crystalline form and (b) reducing this oxide at high temperatures, is entitled to receive a patent on a copending application which claims the same reduction step (b) together with the "preliminary step" of (a) preparing the starting synthetic oxide by a heat treatment which, the prior art demonstrates, is a conventional method for preparing, or providing a synthetic non-magnetic ferric oxide?

2. Whether or not an inventor who has secured a patent on a process including the initial step of heating certain ferric oxide hydrate crystals in air to drive off water of hydration is entitled to receive a second patent including claims to the same process but specifying that the initial heat treatment is carried out for the purpose of changing the magnetic properties of the final product?

3. Whether or not an inventor who has received a patent claiming certain ferric oxide crystals which have an acicular, or elongated, shape and which are adapted to serve as magnetic impulse recording materials, and also claiming a magnetic recording tape utilizing oxide crystals of the same description but without reference to crystal shape, is entitled to receive a second patent on cubic shaped ferric oxide crystals having substantially the same magnetic characteristics on which the patentability of the patent claims was predicated?

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ARMOUR RESEARCH FOUNDATION OF ILLINOIS
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v.

ROBERT C. WATSON, COMMISSIONER OF PATENTS,
APPELLEE.

APPEAL FROM THE JUDGMENT OF THE UNITED STATES DISTRICT COURT FOR THE DISTRICT OF COLUMBIA

BRIEF FOR THE COMMISSIONER OF PATENTS

INTRODUCTION

This is an appeal by Armour Research Foundation of Illinois Institute of Technology from the Judgment (J. A. 38) of the United States District Court for the District of Columbia dismissing its complaint in an action under Section 145, Title 35, of the United States Code, in which appellant sought to have that court authorize the issuance to it of a patent containing claims 35, 36, 37, 38, and 39, of the Marvin Camras application for patent entitled "Improved Coating Material For Magnetic Recording," Serial No. 114,677, filed September 8, 1949.

The Patent Office examiner (Examiner's Answer, J.A. 73), the Patent Office Board of Appeals (Decision, J.A. 81; Reconsideration, J.A. 90) and the District Court (Memorandum Decision, J.A. 36) concurred in holding appellant's claims 35, 36, 37, 38, and 39 here in suit, unpatentable over the claims of the Camras patent No. 2,694,656 (J.A. 93), granted November 16, 1954, to the appellant on an application filed July 25, 1947.

Specifically, method claims 35, 36, 37, and 38 in suit were held by the examiner and the Board to be unpatentable over Camras patent claim 13 in view of each of the publications: Abraham *et al.* (J.A. 100), *Nature*, Vol. 115, No. 2902, page 930, dated June 13, 1925; Welo and Baudisch (J.A. 105), *Philosophical Magazine*, Vol. 50, pages 398 to 408, dated 1925; and Mellor (J.A. 101), *Treatise on Inorganic and Theoretical Chemistry*, Vol. 13, pages 732, 736, 738 and 780, dated 1934. These method claims 35, 36, 37, and 38 were additionally rejected as being unpatentable over claim 21 of the Camras patent. Product claim 39 in suit was held to be unpatentable over product claims 25 and 4 of the Camras patent.

COUNTERSTATEMENT OF THE CASE

The Camras *application in suit* (Specification, J.A. 45 to 55) relates to a process for preparing a finely divided magnetic iron oxide which is intended for use as the magnetic coating material on magnetic sound recorder tapes and to finely divided, magnetic iron oxide particles having, as set forth in claim 39 (J.A. 56), a "coercive force value" (the measure of the ability to retain a state of magnetization, J.A. 17) "between 200 and 450" and a "remanence" "of 200 and above."

The magnetic properties of the iron oxide particles are the result of a reducing treatment, a treatment of a non-magnetic iron oxide (a ferric oxide or a ferrosoferric oxide) with hydrogen or its equivalent at a temparture between 1000° and 1750° F. This reduction treatment is described and claimed in Camras *patent* No. 2,694,656, which was granted on November 16, 1954, to the appellant as assignee. As appellant states (brief, page 10, third complete paragraph) the reduction step which confers the magnetic properties which make the particles desirable for recording use is common to both the *Camras patent* and the *Camras application in suit.* The *application* in suit applies this reduction to an iron oxide, particularly a ferric oxide, Fe_2O_3, a ferric oxide hydrate, $Fe_2O_3 . H_2O$ or a ferrosoferric oxide, Fe_3O_4, which has been exposed to a preliminary step of heating at a temperature between about 1000° and 1750° F. (J.A. 48, first paragraph). This preliminary heating step is described by the Camras *application* in suit (J.A. 54, third paragraph) as being applicable to iron oxides which contain a trivalent iron atom. The appellant's application states that the particles may be "cubic, acicular, scales, irregular grains, etc." The *application* (J.A. 46, last paragraph) also states, "In some application, magnetic iron oxide particles are preferably in the form of cubes." These cubic particles have the chemical composition Fe_3O_4 and are magnetic (J.A. 22, testimony of Marvin Camras; J.A. 48, second paragraph). Claims 37 and 38 in suit (J.A. 55) call for the application of the preliminary heating step to an iron oxide "of cubic particle shape." Claims 35 and 36 in suit include no limitations as to the shape of the treated particles.

Claim 13 of the Camras *patent* (J.A. 98, column 9) includes the preliminary step of "providing a synthetic non-magnetic ferric oxide in crystalline form and of a particle size not over 6 microns in its greatest dimension." Camras patent claim 21 (J.A. 98, column 10) recites the preliminary steps of precipitating crystals of $Fe_2O_3 . H_2O$ (ferric oxide monohydrate) from solution and "heating said crystals in the air to a temperature between 500 and 1550° F., to drive off the water of hydration." Camras patent claim 25 (J.A. 98, column 10) claims particles of magnetic ferrosoferric oxide, or ferric oxide, in the form of elongated crystals of less than 1.5 microns in maximum dimension, which have a coercive force between 245 and 330, and a remanence of above about 500 gauss. Although Camras patent claim 25 specifies that the iron oxide particles be elongated, other claims, including claims 4 (J.A. 97, column 8) and 13, are not so restricted and this absence of specific restrictions as to particle shape is consistent with statements in the specification of the patent (see, for instance, J.A. 94, column 2, lines 44-49).

SUMMARY OF ARGUMENT

1. Claim 13 of the Camras patent, calls for "providing a synthetic non-magnetic ferroc oxide * * *" as the initial step of the claimed "reduction process." This claim, consistent with the specification, is not limited to the treatment of acicular, or needle-like, crystals. The publication of record show that the preliminary heating step on which appellant predicates the patentability of the claims in issue is a conventional method for producing, or "providing", the non-magnetic ferric oxide of Camras patent claim 13. The preliminary step therefore involves no inventive or patentable departure from the process of patent claim 13. Appellant is not entitled to a second patent containing the claims in issue unless the claims disclose invention over and above the claims allowed in the first patent.

2. Claim 21 of the Camras patent recites a preliminary step of heating crystals of ferric oxide monohydrate in the air to a temperature between 500° and 1550° F. The heating step when carried out at 1000° to 1550° F. will result, as shown by appellant's specification and the publication of Welo and Baudisch, in the change to a non-magnetic ferric oxide which is the emphasized feature of appellant's process claims in suit. Accordingly, the circumstance that the patent claim 21 refers to driving off the water of hydration while the process claims in suit refer to ultimate magnetic properties demonstrates no patentable distinction. The reference to cubic particles in claims 37 and 38 in suit involves no basis for patentability since the cubic shape is not attended by

any critical difference of magnetic recording characteristics.

3. Product claim 25 of the Camras patent which claims a ferromagnetic iron oxide having a coercive force and particle size comparable to those called for in product claim 39 of appellant's application, and a remanence value higher than that called for in application claim 39 is specific to particles of acicular, or needle-like, shape but product claim 4 of the Camras patent is not limited to magnetic iron oxides of any particular shape. Consequently, claim 39 in suit which calls for cubic shaped particles which have no properties or use characteristics which are critically different from, or superior to the claimed particles of the Camras patent, defines no patentable invention over the Camras patent claims.

ARGUMENT

1. As the District Court recognized (J.A. 37 and 38) the Patent Office examiner (J.A. 75) and the Board Of Appeals (J.A. 83) found that no invention would be involved in employing conventional prior art practices for "providing a synthetic non-magnetic ferric oxide in crystalline form and of a particle size not over 6 microns * * *" as called for in claim 13 of the Camras patent (J.A. 98). As appellant's brief concedes (page 10, third complete paragraph) claim 13 of the patent "may be broad enough to read upon the new improvement since it is directed essentially to the reduction, and that step is common to both processes." The record includes no serious challenge to the Patent Office holding that the secondary refer-

ences Welo and Baudisch (J.A. 105), Abraham (J.A. 100) and Mellor (J.A. 101) show the preparation of the non-magnetic ferric oxide (the alpha form) from a magnetic iron oxide having a trivalent iron atom by heating the magnetic iron oxide in air at temperature above approximately 550° C. (1022° F.). [Welo and Baudisch, for instance, on page 401, first complete paragraph, of their article (J.A. 106), succinctly describe the heat transformation to the non-magnetic ferric oxide.] Indeed, appellant's brief (page 13, second complete paragraph) contains the admission that the cited secondary references "merely show that it is old to demagnetize iron oxide material by high temperature heat treatment."

In view of the disclosures in the secondary references of a conventional procedure for preparing the non-magnetic ferric oxide by high temperature treatment, it is submitted that the "preliminary" step upon which appellant predicates invention was apparent and obvious. The uninventive preliminary step provides no valid basis for a second patent since, as the decisions of the U. S. Court of Customs and Patent Appeals and this Court demonstrate, in order for a second patent to issue the claims of an application must differ inventively from the claims of a co-pending patent issued to the same applicant. *In re Isherwood*, 46 App. D. C. 507; *In re Copeman*, 30 C.C.P.A. 962, 135 F. 2d 349; *Kistler* v. *Coe*, 79 U. S. App. D. C. 36, 142 F. 2d 94; *In re Asseff*, 36 C.C.P.A. 867, 173 F. 2d 253; *In re Ockert*, 44 C.C.P.A. 1024, 245 F. 2d 467. In determining whether the claims of an application are patentably distinct from the claims of the same applicant's copending patent it is proper to consider what is disclosed in the prior art. *In re Ockert, supra; In re Barge*, 25 C.C.P.A. 1058, 96 F. 2d 314; *In re Mason*, 20 C.C.P.A. 782, 62 F. 2d 185; *In re Isherwood, supra.*

Appellant's brief (pages 10 and 11) apparently attempts to give "providing a synthetic non-magnetic ferric oxide," as employed in Camras patent claim 13, a restrictive meaning which excludes the "preliminary" step of the application claims 35, 36, 37, and 38. It is not seen how restricting the meaning of "providing" would do appellant any service because the question involved is not the scope of the limitations that can be imported into term "providing" but whether the "preliminary" step of process claims in issue amounts in fact to an inventive departure from the process claims of the patent, and specifically claim 13. Therefore, even if the step of "providing" in the patent claim 13 had been replaced by a mere designation of the starting material for the reduction step the preparation of that starting material by a procedure shown to be conventional would not amount to an inventive improvement on the patented process.

Appellant's arguments, moreover, do not support appellant's restricted interpretation for the "providing" step of Camras patent claim 13. Appellant's brief (page 11, third paragraph), for instance, suggests that "providing" means the step of producing "initial" crystals from solution, "*or an equivalent* of such step" (emphasis added). Appellant's brief in the same paragraph compounds the inconsistency by urging that "providing a synthetic non-magnetic ferric oxide" as employed in patent claim 13 is "merely the statement of an *existing initial condition* and not an *instruction* to the art that an already magnetic material should be demagnetized." This contention ignores not only appellant's concession that "providing" refers to a "step" but also the identification of the crystals as "synthetic." Obviously a "synthetic" non-magnetic ferric oxide must be manufactured.

The decisions cited in appellant's brief, *Research Products Co.* v. *Tetrolite Co.* (C.C.A. 9, 1939), 106 F. 2d 530, 535, (appellant's brief, page 13), *Hope Basket Co.* v. *Products Advancement Corp.* (C.A. 6, 1951), 187 F. 2d 1008, 1012, and *Oliver United Filters, Inc.* v. *Silver* (C.A. 10, 1953), 206 F. 2d 658, (appellant's brief, page 14), are not believed to demonstrate appellant's right to receive two patents on copending applications which do not in fact claim two separate inventions. *Research Products Co.* v. *Tetrolite Co., supra,* which is said by appellant (brief, page 13) to present an "identical" fact situation, involved the patentability of a process for breaking oil emulsions over the patentee's copending patent to a process of water purification, the two processes using different treating compounds so that the claims of the two patents, unlike the patent and application of appellant, involved different treating materials, different treatments, and different objectives. *Hope Basket Co.* v. *Products Advancement Corp., supra,* involved two "essentially distinct and separable" inventions, a basket making machine in one patent, and a basket which could be made by hand or by other processes without infringing the claims of the first patent in the other patent.

The court's statement in *Oliver United Filters, Inc.* v. *Silver* (C. A. 10, 1953), 206 F. 2d 658, "if the second Silver patent defines a process or apparatus different from the discoveries of the first patent, it does not constitute double patenting even though it relates to the same invention," would seem to suggest that the court believed that the claims of two copending patents of the same inventor need not be inventively distinct, but such a ruling would be contrary to such decisions as *Kistler* v. *Coe,* 79 U. S. App. D. C. 36, 142 F. 2d 944; *Weatherhead Company et al.* v. *Drillmaster Supply Company et al.*, (C.C.A. 7, 1955), 227 F. 2d 98; *In re Isherwood,* 46 App. D. C. 507, and numerous decisions in the U. S. Court of Customs and Patent Appeals exemplified by that court's decisions cited above. It is notable that the court in *Oliver United Filters, Inc.* v. *Silver, supra,* included in the decisions on which it relied, *In re Thompson,* 20 C.C.P.A. 725, 62 F. 2d 90, and *In re Peiler,* 19 C.C.P.A. 1051, 56 F. 2d 878. *In re Thompson,* however, asserted its reliance on *In re Isherwood, supra,* and stated the question of patentability as follows:

> "The sole question, therefore, as to appellant's claims 5 and 7 is whether they present anything inventive over his patent claims."

Similarly, the court in *In re Peiler, supra,* made the following holding:

> "The appellant, having disclosed or claimed no matter patentably distinct from that claimed in his existing patent, can not have a second patent upon the same invention."

The decision of the U. S. Court of Customs and Patent Appeals in *In re Ward and Switzer*, 43 C.C. P.A. 1007, 236 F. 2d 428, provides a recent illustration of the principle that mutual "cross-reading" of the claims of an application and patent is not indispensable to a holding of double-patenting. This decision, which held it axiomatic that there must be two inventions present if two patents are to be granted, refused a patent to the joint inventors on a claim to a process of testing castings for flaws which used a color contrasting testing agent where the sole patent of one of these inventors claimed the use of a luminescent agent in the same process. The present Court in *Switzer* v. *Watson* (Appeal No. 13948) —— U. S. App. D. C. ——, 251 F. 2d 386, refused to overturn a similar rejection of Switzer's application claim 11, which claimed the use of a testing agent having an oily vehicle with a dye dissolved therein, over the claims of the same Ward patent which was the basis for rejection in *In re Ward and Switzer, supra.*

2. The Board of Appeals in its decision (J.A. 81) affirmed also the rejection of the appellant's claims 35 to 38 as being unpatentable over claim 21 of the Camras patent. Since patent claim 21 (J.A. 98) claims the preparation of the non-magnetic ferric oxide for the subsequent reduction step by "heating said crystals [$Fe_2O_3 . H_2O$] in the air to a temperature between 500 and 1550° F. to drive off the water of hydration" it is not apparent that the preliminary heating or oxidation at temperatures between 1000° and 1750° F. specified in the application claims provides a patentable distinction. The explanation that the heating to drive off the water of hydration when

carried out at the high temperatures suitable for this operation (J.A. 2) accomplishes a desirable result apart from the dehydration is in the nature of merely a scientific explanation of what takes place in a previously claimed process, and as such provides no basis for patentability. *In re James*, 23 C.C.P.A. 1124, 83 F. 2d 313.

Appellant's argument (appellant's brief, page 12) that patent claim 21 is directed to the treatment of acicular, or needle-like, particles rather than cubic particles ignores the fact that the Camras application describes the claimed preliminary treatment as applicable not only to cubic particles but to "acicular, scales, not only to cubic particles but to "acicular, scales, irregular grains, etc." (J.A. 54, second complete paragraph) and includes in specific Examples VII and VIII (J.A. 52) a description of the preliminary treatment and final reduction of "practically non-magnetic hydrous ferric oxide powder." These disclosures, especially when coupled with the application's statement (J.A. 47, last paragraph) that the claimed heating and reduction steps are "particularly applicable to the treatment of ferric oxide, Fe_2O_3, its hydrate, $Fe_2O_3 . H_2O$, or to ferrosoferric oxide, Fe_3O_4." (emphasis added), would seem to rebut any contention that the reference to cubic particles in the application claims would be of patentable significance. In fact, the lack of patentable significance of the reference to "cubic" particles in claims 37 and 38 in suit is indicated by the *absence of any limitation as to shape in claims 35 and 36 in suit.* Since the appellant has not suggested that the iron oxide particles of a cubic shape have magnetic properties which are critically different from those possessed by particles of other shapes it is not apparent why the recitation of "cubic" shape in claims 37 and 38 in suit makes these claims patentable over patent claims which cover particles of a shape originally considered to be preferable or particles without limitation as to crystalline shape.

3. The evidence of record, it is submitted, provides no basis for the allowance of the appellant's application claim 39 merely because it calls for a *cubic* particle shape rather than the elongated particle shape mentioned in Camras patent claim 25. This difference, as the Board's decision on reconsideration (J.A. 89) held, does not warrant grant of another patent, on the basis of claim 39 in suit. The patent claims like the specification on which they are based are not restricted to cover only the elongated, or acicular, crystals. Product claims 4 and 5 (J.A. 97), for instance, of the patent are directed to a magnetic record member in which the synthetic iron oxide, which is the novel feature of the claimed magnetic record member, is not restricted as to its crystalline shape.

If the iron oxides in cubic crystalline form have any magnetic-recording advantages not possessed by the acicular crystals the record does not show these advantages. The Camras patent claims treated the various crystalline forms of its magnetic iron oxides as being all suitable for magnetic recording purposes (claims 4, 5, and 13) and pointed to (claim 25) a preferred acicular form. The Camras application (J.A. 54, second paragraph) is similarly non-restrictive as to the shape of the particles. No invention, therefore, is apparent in a decision by the patentees to seek a new patent monopoly upon a cubic form having recording characteristics which are desirable but not critically different. Presumably, if appellant were granted a patent on the basis of the cubic shape of the particles it could seek and obtain in subsequent applications successive patent monopolies on the scale-like crystals, the irregular crystals, and those of other shapes without showing that the shape of the particles involves inventive significance. It is not believed that the patent laws favor such a policy of *monopoly by attrition.*

CONCLUSION

It is respectfully submitted that, for the reasons stated hereinabove, the conclusions reached by the Patent Office tribunals and the District Court that the claims in suit are unpatentable over the claims of the Camras patent No. 2,694,656 are correct and have a reasonable basis in the record, and that under the standards of appellate review set forth in *Abbott et al.* v. *Coe*, 71 App. D. C. 195, 109 F. 2d 449 and *Standard Oil Development Co.* v. *Marzall*, 86 U.S. App.D.C. 210, 214, 181 F. 2d 280, 284, the decision of the District Court should be affirmed.

Respectfully submitted,

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OCTOBER, 1958

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REPLY BRIEF FOR APPELLANT.

In the
United States Court of Appeals
DISTRICT OF COLUMBIA CIRCUIT.

Appeal No. 14,550.

United States Court of Appeals
For the
District of Columbia Circuit
FILED OCT 23 1958
Joseph W. Stewart
CLERK

ARMOUR RESEARCH FOUNDATION OF ILLINOIS
INSTITUTE OF TECHNOLOGY,
Appellant,
vs.
ROBERT C. WATSON, Commissioner of Patents,
Appellee.

Appeal from the Judgment of the United States District Court for the District of Columbia.

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In the

United States Court of Appeals

DISTRICT OF COLUMBIA CIRCUIT.

Appeal No. 14,550.

ARMOUR RESEARCH FOUNDATION OF ILLINOIS
INSTITUTE OF TECHNOLOGY,
Appellant,
vs.
ROBERT C. WATSON, COMMISSIONER OF PATENTS,
Appellee.

Appeal from the Judgment of the United States District Court for the District of Columbia.

REPLY BRIEF FOR APPELLANT.

This brief is necessitated by certain misstatements of fact and by the erroneous conclusions drawn therefrom appearing in Appellee's Brief.

SUMMARY OF THE CASE.

This action, brought under Section 145, Title 35, of the United States Code, seeks to compel the Commissioner of Patents to issue a patent to Marvin Camras, as assignor to the Plaintiff-Appellant.

The invention on which a patent is sought is directed to the manufacture of magnetic iron oxides for magnetic record members.

Iron oxides suitable for modern magnetic recording purposes are not to be found in nature. All present-day magnetic oxides are manufactured from synthetically prepared oxides of iron, *i. e.,* synthesized compounds of iron and oxygen. Such compounds are, of course, well known chemical entities, but their magnetic properties vary tremendously. For modern magnetic recording purposes, such compounds must have a unique combination of magnetic properties—a moderately high "coercive force", or ability to resist demagnetization, and a high "remanence", or ability to record a wide spectrum of sound frequencies.

In his prior patent, No. 2,694,656, Mr. Camras demonstrated that the properties desired for magnetic recording purposes could be achieved by treatment of the synthetically prepared iron oxide material in a "reducing" atmosphere, such as hydrogen gas, at elevated temperatures. When this process is applied to the acicular or needle-like form of synthetic iron oxide preferred by the patent, the process suffices *without other treatment* to impart to the material a coercive force between 200 to 450 and a remanence of 200 and above.

Quite apart from the invention of his Patent No. 2,694,656, but while that patent was pending, Mr. Camras discovered that other iron oxide starting materials such as those of cubic particle form which respond somewhat, although not so well as the acicular form, to the reduction treatment, could be provided with the desired properties for magnetic recording by preceding the reduction treatment with a high temperature (1000° to 1750° F.) heat treatment.

There is no known reason why the preliminary heat treatment step should render the subsequent reduction step effective, but, as the method claims here specifically point out, without such step the high coercive force values desired cannot be obtained with certain starting oxides.

POINTS OF ERROR IN APPELLEE'S BRIEF.

On Page 3, Appellee's Brief indicates that the reduction treatment of the issued patent is carried out at temperatures between 1000° and 1750° F. This statement is apparently an inadvertent error, since the patent specifies a reduction temperature between 500° and 1000° F. (J. A. 95, Column 2, lines 31-35).

On page 4 of the brief, the statement is made that "Claims 35 and 36 in suit include no limitations as to the shape of the treated particles." This statement is literally true, but it overlooks the express language of claim 35 which states that the material, through the effects of the combined high temperature and reduction treatments, achieves a coercive force "* * * *higher than could be obtained by employing the same reducing conditions in the absence of said preliminary heating step.*" Claim 36, being dependent upon claim 35, carries the same limitation. Thus, the claims by their very terms *exclude* extremely reactive materials, such as those of acicular form, which do not require the high temperature pre-treatment for attaining the required coercive force properties.

Appellee contends that Appellant is seeking another patent on the same subject matter on which it received Patent No. 2,694,656. Its argument follows these lines: Since the high temperature pre-treatment forming an essential step in the new process serves to demagnetize the iron oxide material if it is already somewhat magnetic, and since the issued Camras patent teaches one how to convert non-magnetic material into material suitable for recording purposes, the newly presented claims to the combination of steps and the patented claims are for one and the same invention. In support of this argument, Appellee's Brief (p. 4) relies upon claim 13 of the issued patent which includes the introductory step of "providing a synthetic non-magnetic ferric oxide * * *." The argument overlooks the fact, uncontroverted by Appellee, that the patent specification (J. A. 94-97) describes the provision of a starting material for the reduction process which is *never* magnetic prior to the reduction process.

Appellee also relies upon claim 21 of the issued patent, which recites a process for the treatment of the compound $Fe_2O_3.H_2O$, by dehydrating that compound at a temperature between 500 and 1550° F. prior to the reduction treatment. Again, the argument ignores the fact, uncontroverted by Appellee, that the hydrated material is inherently non-magnetic, with or without dehydration, and unlike the starting material of the new application is acicular and therefore inherently responds *to reduction alone* to achieve the desired magnetic properties.

Appellee's argument on double patenting cannot withstand critical analysis. The claims of the issued patent and those being urged here do not embrace the same inventive concept. At the expiration of the issued patent, the public will be free to use the patented process on all non-magnetic synthetic iron oxides which, *by reduction alone,* achieve the designated magnetic properties. The claims here in issue are directed solely to the use of (1) a material which does not respond adequately to reduction alone to achieve ideal magnetic properties; *and* (2) to the processing of such starting material by *a combination* of a high temperature pretreatment step with the reduction step. The two inventive concepts are thus not the same and a patent granted on the new invention would not be a duplicate of that already granted.

Appellee also pieces together the steps of the new process in the following way. Others have shown that iron oxides could be demagnetized by high temperature treatment; Camras in his prior patent taught how to make good magnetic material from non-magnetic material; ergo, there is no invention in using a treatment known to demagnetize iron oxides and then follow that with the reduction step of the Camras patent. Application of elementary logic, if nothing else, indicates the fallacy of this argument. Where is the missing link in this syllogism? Where is there any suggestion prior to Mr. Camras' present invention that a step known to demagnetize iron oxides could be instrumental in making weakly magnetic materials *more* magnetic upon subsequent reduction? We do not find any such suggestion in the evidence presented at trial, nor do we find it from Appellee's analysis of the prior art.

Kistler v. *Coe,* cited by Appellee, is inapposite. Kisler had obtained a patent on an aerogel, *per se.* He then tried to obtain another patent on a method of insulating buildings which involved nothing more than using his new aerogel in the otherwise old construction method. This Court clearly indicated that Kistler's inventive concept had been exhausted by the issuance of the patent, and he was not entitled to a second patent. This fact situation is entirely different from that presented here where the patent applicant has provided an *improvement* over a broader prior patent.

In *Weatherhead Co.* v. *Drillmaster Supply,* plaintiff had a patent on a joint for pipe connections and a later patent on a metal packing ring for use with such a joint. The court found that an infringement of one patent was necessarily an infringement of the other, so held double patenting to exist. No such situation exists here.

In re Isherwood, relied on by Appellee, was a case in which the Court held one *species* of an invention unpatentable over another *species*. There was no question presented of a *new combination* of process steps being patentable over a disclosure of the steps in unrelated parts of the prior art.

In re Thompson, was a case involving a rejection of apparatus claims upon an issued method patent to the same inventor. No question of a new combination of method steps was involved.

In re Peiler involved the patentability of claims in a later application directed to the *identical subject* matter patented to the applicant previously, but rephrased. Clearly, this fact situation is not present here.

In re Ward and Switzer involved, as the Court found, a single inventive concept relating to a method of flaw detection. The patentability of substituting one species of dye for another was the sole issue on the question of double patenting.

Again, in the cited case, there was no question of the patentability of a combination of method steps not claimed in the prior patent.

CONCLUSION.

Appellants respectfully submit that the claims here in issue patentably distinguish from those of the Camras patent by excluding from their scope those starting iron oxides that are sufficiently reactive to acquire the desired magnetic properties by reduction, alone, without any such preliminary high temperature conditioning step as here claimed in combination with the reduction step.

Appellants further submit that by virtue of so excluding the reactive iron oxides of the patent, the present claims are not bad because of double patenting. The public, *after* the expiration of the Camras patent, will be free to practice the invention of that patent without fear of infringing any of the claims here sought, if patented. The essential element of double patenting is entirely absent here.

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JOINT APPENDIX

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

United States Court of Appeals
For the
District of Columbia Circuit
FILED AUG 12 1958
Joseph W. Stewart
CLERK

No. 14,550

ARMOUR RESEARCH FOUNDATION OF ILLINOIS
INSTITUTE OF TECHNOLOGY,

Appellant,

v.

ROBERT C. WATSON,
Commissioner of Patents,

Appellee.

APPEAL FROM THE UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF COLUMBIA

Washington, D. C.
ROBERT I. THIEL
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EX 3-0825

UNITED STATES COURT OF APPEALS
For The District Of Columbia Circuit

No. 14,550

ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY,

Appellant,

v.

ROBERT C. WATSON,
Commissioner of Patents,

Appellee.

APPEAL FROM THE UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF COLUMBIA

JOINT APPENDIX

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51 [Filed May 17, 1956]

IN THE DISTRICT COURT OF THE UNITED STATES
FOR THE DISTRICT OF COLUMBIA

ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY, Chicago, Illinois. Plaintiff,	
vs.	Civil Action No. 2084-56
ROBERT C. WATSON, Commissioner of Patents, Washington, D. C., Defendant.	

COMPLAINT FOR ISSUANCE OF PATENT

1. Plaintiff, ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY, is a not-for-profit corporation organized under the laws of the State of Illinois, having an office and place of business at 10 West 35th Street, Chicago 16, Illinois, and is the owner by assignment of the entire right, title and interest to the hereinafter described invention and patent application of MARVIN CAMRAS.

2. Defendant, ROBERT C. WATSON, has his official residence in the District of Columbia, and is here being sued in his official capacity as Commissioner of Patents of the United States.

3. This action arises under U.S.C., Title 35, Section 145.

4. Marvin Camras is the inventor of an invention embodying improvements in an IMPROVED COATING MATERIAL FOR MAGNETIC RECORDING. On September 8, 1949, said Marvin Camras duly made application for United States Letters Patent on said invention, said application being assigned Serial No. 114,677, and said application is not abandoned.

52 By lawful instrument in writing, said Marvin Camras has assigned said invention and said application to Plaintiff, ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY, and Plaintiff is still the lawful owner of said invention and said application.

5. Said application was duly prosecuted, and was passed upon by

the Primary Examiner in the Patent Office without the allowance of any claims.

6. Plaintiff and said Marvin Camras, through their attorneys, appealed from the decision of the Primary Examiner to the Patent Office Board of Appeals, on the basis of the following claims:

> 35. In a method of making permanent magnet material from a starting synthetic iron oxide containing a trivalent iron atom having a coercive force less than 200 oersteds and having a particle size of one micron and less by reducing said iron oxide at an elevated temperature, the improvement which comprises the preliminary step of heating such starting synthetic oxide to a temperature between 1000° and 1750° F. for a sufficient length of time that when subsequently reduced at an elevated temperature below that used in said preliminary heating step said iron oxide acquires a coercive force value in excess of 200 oersteds and higher than could be obtained by employing the same reducing conditions in the absence of said preliminary heating step.
>
> 36. The method defined by claim 35 in which the preliminary heating step is carried on in the presence of an oxidizing atmosphere, and the subsequent reduction is carried out in a reducing atmosphere at a temperature of between 400° and 1200° F.
>
> 37. The method defined by claim 35 in which the starting synthetic oxide is of cubic particle shape, the preliminary heating step is carried out in the presence of air, the subsequent reduction step is carried out in a hydrogen atmosphere and the resulting reduced iron oxide has a coercive force value in excess of 300 oersteds.
>
> 53 38. The method defined by claim 35 in which the starting synthetic oxide is ferrosoferric oxide (Fe_3O_4) of cubic particle shape, the preliminary heating step is carried out in the presence of air, the reduction step is carried out in a reducing atmosphere and the resulting reduced iron oxide is re-oxidized at a temperature from about 400° to 900° F. in an oxidizing atmosphere to

produce an iron oxide having a coercive force value in excess of 200 oersteds.

39. Permanent magnet material adapted to form an element of a magnetic impulse record member consisting of synthetic magnetic iron oxide particles of a cubic particle shape less than one micron in size and having a coercive force value (H_c) between 200 and 450 and a remanence (B_r) of 200 and above.

7. The Patent Office Board of Appeals, in a decision rendered March 8, 1956, affirmed the decision of the Primary Examiner with respect to the rejection of the above quoted claims. Within the 30 day period specified in Rule 304 of the Patent Office Rules of Practice, on March 26, 1956, Plaintiff's attorneys filed a Petition for Reconsideration with the Patent Office Board of Appeals. In a decision rendered April 18, 1956, in response to said Petition, the Patent Office Board of Appeals again affirmed the decision of the Primary Examiner as to the rejection of the above quoted claims. Profert of a duly authenticated copy of said application and of the foregoing proceedings in the Patent Office involving said application is hereby made. No appeal has been taken from said decisions of the Patent Office Board of Appeals to the United States Court of Customs and Patent Appeals, and no notice of such an appeal has been given to the Commissioner of Patents.

8. During the pendency of the aforementioned patent application Serial No. 144,677 in the Patent Office, the Plaintiff, as assignee of the said Marvin Camras, was prosecuting in the United States Patent Office a second patent application entitled "PERMANENT MAGNET
54 MATERIAL AND METHOD OF MAKING THE SAME", said application bearing Serial No. 763,486 and having been filed on July 25, 1947. Said application Serial No. 764,486 ultimately issued as United States Letters Patent No. 2,694,656 on November 16, 1954 and Plaintiff, ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY, is still the lawful owner of said Letters Patent.

9. The Commissioner of Patents, by the Patent Office Board of Appeals, contends that claims 35 to 38 inclusive should be rejected

as unpatentable over claim 13 of Camras United States Patent No. 2,694,656 in view of certain publications as follows:

Abraham et al, "Nature", page 930, Vol. 115, No. 2902, June 13, 1925;

Welo and Baudisch, "Two Stage Transformation of Magnetic into Hemotite", Phil. Mag., Vol. 50, pages 399-408 (1925);

Mellor, "Treatise on Inorganic and Theoretical Chemistry", Vol. 13, pages 732, 736, 738, 380, Pub. Longmans, Green & Co., New York City, 1934.

The Commissioner of Patents, by the Patent Office Board of Appeals, further contends that claims 35 to 38, inclusive, should be rejected as unpatentable over claim 21 of the aforementioned Camras Patent No. 2,694,656. The Commissioner of Patents further contends that claim 39 should be rejected as unpatentable over claim 25 of the aforementioned Patent No. 2,694,656.

10. Plaintiff avers that the aforementioned Camras Patent No. 2,694,656 was improperly applied by the Commissioner of Patents in the rejection of the claims 35 to 39 inclusive and that any proper application of the aforementioned Patent No. 2,694,656 alone or in combination
55 with any or all of the aforementioned technical articles fails to anticipate the said claims 35 to 39 inclusive, and that said claims 35 to 39 inclusive properly define patentable subject matter thereover.

WHEREFORE, Plaintiff prays that the Commissioner of Patents be directed to grant a patent containing said claims 35 to 39 inclusive, and such other claims as upon hearing this Court may find proper for adequate protection of said invention, and for such further relief as equity may require.

/s/ Benjamin H. Sherman

/s/ Otto R. Krause

Hill, Sherman, Meroni, Gross & Simpson * * *
Attorneys and Counsel for Plaintiff
/s/ Smith Michael & Gardiner
Smith, Michael & Gardiner * * *
Of Counsel

56 [Filed July 3, 1956]

Clarence W. Moore, Solicitor,
United States Patent Office,
Washington 25, D. C.,
Attorney for Defendant.

ANSWER TO THE COMPLAINT

To the Honorable the Judges of the United States District Court for the District of Columbia.

1, 2, 3, 4, 5, 6, 7, 8, 9. The defendant admits the allegations of paragraphs 1 to 9, inclusive, of the complaint.

10. The defendant denies the allegations of paragraph 10 of the complaint.

FURTHER ANSWERING, the defendant states that the plaintiff properly was refused a patent on the application of Marvin Camras, Serial No. 114, 677, containing any of claims 35, 36, 37, 38 and 39 thereof, for the reasons given and in view of the references cited in the Examiner's Answer and the decisions of the Board of Appeals in said
57 application. Profert of copies of said references, answer and decisions hereby is made.

Respectfully submitted,

/s/ C. W. Moore

Solicitor, U. S. Patent Office,
Attorney for Defendant.

July 2, 1956.

[Certificate of Service]

1 [Filed June 25, 1958]

Washington, D. C.,
January 15, 1958.

The above-entitled action came on for trial on the merits, before the HONORABLE JOSEPH C. McGARRAGHY, United States District Judge, at 10 o'clock a.m.

APPEARANCES:

On behalf of the Plaintiff:

WILLIAM A. SMITH, JR., Esq.
BENJAMIN H. SHERMAN, Esq.
OTTO R. KRAUSE, Esq.

On behalf of the Defendant:

ARTHUR H. BEHRENS, Esq.

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3

PROCEEDINGS

DEPUTY CLERK: Armour v. Watson.

MR. SMITH: May it please the Court, I appear before Your Honor on behalf of the Armour Research Foundation of Illinois and respectfully request that Mr. Otto Raymond Krause of Chicago, Illinois, who is not a member of the bar of this court but is a member of the bar of the highest court of the State of Illinois, be permitted to present this case.

THE COURT: Mr. Krause, I will be very glad to have you do so.

MR. KRAUSE: Thank you, sir.

MR. SHERMAN: If Your Honor please, we have a trial brief that we would like to hand up to you. I have given a copy to our opponent.

This action comes before Your Honor under Section 145, Title 35 of the United States Code, and is a mandamus action to direct the Commissioner of Patents to issue a patent to one Marvin Camras, assignor to the Armour Research Foundation of the Institute of Illinois. The Armour Research Foundation, as it is called commonly, is a non-profit organization, a corporation of Illinois, and the owner of the Camras application in this suit.

Mr. Camras, the inventor, will be a witness here, He is a world-renowned scientist in the field of magnetic recording, as Your Honor is doubtless familiar with.

4 THE COURT: Let me disabuse your mind, I am not too familiar with it. I would like to be made more familiar with it.

MR. SHERMAN: All right, Your Honor. The art of recording magnetically both sound and other impulses has developed very greatly in the last twenty years. Actually, the art has its origin in the work of a Danish scientist named Poulsen who, at about the turn of the century, discovered that certain magnetic wires when subjected to impulses would change their magnetic properties and these changed magnetic properties could then be used in reproducing the sound or other impulse that initially caused the change in the magnetic properties.

The work of Poulsen on wires resulted in practically no commercially successful devices. It was largely a laboratory experiment and it was not until the '30's that the recording on magnetic wires and magnetic tapes was resumed.

The recording on wire is not so satisfactory, for the reason that in playing back the recording the wire, which is of very fine diameter, is apt to twist, turn, and otherwise become distorted, so it soon became evident that a better recording medium would be a tape.

Now, the tape traditionally has a nonmagnetic backing or carrier for the magnetic particles. The backing may be paper or plastic and the coating of the magnetic particles is applied to the paper and forms what is called a sound track and it is the magnetic particles that assume or
5 take on the effects of the impulses that are caused by the transmission of the voice through the mechanism to the tape. Now, the magnetic particles, as early developed, did not have desirable properties for a magnetic impulse record member in that they did not have what we term a sufficiently high coercive force or retentivity of the magnetism to be rather permanent records.

The tapes are used not only in voice and music recording but also to record signals for industrial processing control, so that they have to have retentivity of magnetism over a rather lengthy period of time.

Now, in Mr. Camras' first patent on magnetic powder and tape, and the method of making the powder, the solution for the first time was made of the relationship between the physical and the magnetic properties best suited for magnetic recording. That patent has been cited against the present application. In the patent, Mr. Camras found that the physical shape best adapted for the purpose was the acicular shape of particle, by which is meant a needle-like one that is much longer than it is thick or wide. He found that by simply precipitating the acicular particles and growing the crystals to a favorable length, he could then take these which were non-magnetic particles and subject them to process steps that developed the most desirable magnetic properties for recording purposes.

6 Those steps include a reduction step, either by hydrogen or other reducing gas, and then, if desired, a following oxidation step. The particles then had the combined magnetic properties and physical properties that best adapted them for magnetic recording purposes. That patent which we call the '656 patent, has been licensed and is now in commercial use to a very considerable extent. The Minnesota Mining people are on the license, and there are others who are making the tape under the issued Camras patent.

In the course of his work during the development of the subject matter of his patent, Mr. Camras found that there were other particles other than acicular in form that did not respond to the simple steps of reduction and oxidation to develop these magnetic properties and, while the '656 patent was pending, he filed the instant application involved in this suit; and this application is directed to a pre-treatment of particles that do not, by the simple reduction or reduction followed by oxidation steps, develop the desired magnetic properties.

The particles in the present application are generally cubic in shape or equant, that is, having about equal dimensions in all directions. They are also extremely fine particles and it is these particles that form the subject matter of the application here before Your Honor.

7 These particles do not respond, as I have said, to the simple steps

of reduction or reduction followed by oxidation in accordance with the issued patent process. Consequently, it was thought desirable to file this other application which is before Your Honor and which was copending with the patent.

In the pending application, the novel step that involved a distinctly different concept from any step disclosed in the '656 patent or claimed in that patent is the step of pre-heating the particles to a temperature of between about 1000° to 1750° F. Within that temperature range, the particles, although initially feebly magnetic or somewhat magnetic, lose their magnetism entirely. There is a known phenomenon that covers that and that is what is called the Curie point; if magnetic materials are heated above their Curie point, they then lose their magnetism.

Now, strange as it may seem, considering the fact that these cubic particles, the small particles, frequently do have some magnetism, Mr. Camras found that by pre-heating the particles until they lost their magnetism, he could then subject the pre-heated particles to the simple steps of the issued patent of either reduction or reduction followed by oxidation and obtain a product that was just as satisfactory, in terms of its magnetic properties, as the particles of his issued patent.

There is no explanation for it. The art knew that particles could
8 be heated above the Curie point and lose their magnetism, and such art is relied upon by the Patent Office. But what the references relied upon by the Patent Office do not show is that there is any correlation between this step of heating to above the Curie point and the development in the subsequent particles of the desired magnetic properties for recording purposes.

The Patent Office Examiner and the Board of Appeals rejected the claims of the present application on a doctrine akin to double patenting. What the Patent Office and the Board of Appeals said, in effect, was that by your issued patent you have claimed subject matter from which the present subject matter of this application differs in no inventive sense. In the issued patent, there is one claim that includes a step of heating to drive off water of hydration. In acicular particles, the common method and that disclosed in Mr. Camras' patent was to precipitate the acicular particles from solution and, when so precipitated, the particles had water of hydration, that is, water came out as a loosely bound part of the crystalline structure, and that water of hydration could be driven off separately. And one of the claims to which the Board of Appeals refers is a claim that covers the separate driving off of the water of hydration from the acicular particles before the treatment of those particles to develop the magnetic properties of the particles.

9 Now, it is not necessary, and the issued patent clearly shows that it was not necessary, to drive off the water of hydration. The hydrated particles could be subjected to the steps of reduction or of reduction followed by oxidation without a preliminary step of driving off the water of hydration, and the single claim that shows or claims the driving off of the water of hydration specifically indicates that is the function of the initial heating step, but the Patent Office has seized upon that as being a step analogous to the heating step in the instant application to precede the reduction and oxidation steps.

In the instant application, as I have tried to point out, the preheating step is necessary in order that the cubic and fine particles develop the desired magnetic properties. In the issued patent, it is not necessary and such heating as was shown in the issued patent was solely for the purpose of driving off water of hydration so that in the subsequent reduction step, it was not necessary to carry out the reduction either at a temperature or for a length of time sufficient to drive off the water of hydration. The reduction step could then be carried out merely under the conditions of time and temperature necessary to effect the reduction, provided the particles had first been dehydrated.

The other claim of the patent to which the Patent Office refers, is the claim that has to do with providing a non-magnetic iron oxide. Well,

10 the word "providing" has to be construed in the light of the issued patent to mean the provision in accordance with that method and that method of the issued patent included the precipitation from solution of

the iron oxide that formed the starting point of the heat treatment steps of the patent.

Now, in the prior art, the Patent Office has seized upon art that shows the heating to above the Curie point to produce a non-magnetic iron oxide and the Patent Office has taken the view that that is one of the ways that the claim of the patent covers in providing a non-magnetic iron oxide, but that is a distortion of the language and not in accord with the disclosure in the specification of the issued patent because, in the present application, the step of pre-heating the iron oxide of cubic or very fine particle form is essential and without it, the cubic particle shaped particles cannot develop the desired magnetic properties. With it, the cubic particle shaped particles can develop the same magnetic properties to the same degree and of the same effectiveness as in the issued patent.

But the claims of the issued patent do not include this pre-heating step of the present application and we submit that the present application, therefore, brings out a different and distinct inventive concept from that of the issued patent, and in reference to the issued patent on the question of double patenting, only the claims of the issued patent may be considered.

11 I have referred to the specification of the issued patent solely for the purpose of construing the term "providing" as used in one of the claims and that has a precedent in the authorities.

I should like at this time to offer the certified copy of the file wrapper and contents of the application in suit as Plaintiff's Exhibit No. 1 and ask that it be so marked.

DEPUTY CLERK: Plaintiff's Exhibit No. 1.

THE COURT: It may be received.

(Certified copy of file wrapper in Camras Application Serial No. 114,677, was marked Plaintiff's Exhibit No. 1 and received in evidence.)

MR. SHERMAN: That is all.

MR. BEHRENS: If Your Honor please, I would like at this time to offer in evidence the folder of exhibits for the Patent Office, representing the Decision of the Board of Appeals and the references upon which it is based, the Examiner's Statement, and the Board's Decision on Reconsideration.

DEPUTY CLERK: Defendant's Exhibit No. 1.

THE COURT: It may be received.

(Patent Office folder of exhibits was marked Defendant's Exhibit No. 1 and received in evidence.)

MR. BEHRENS: The Board of Appeals and the Examiner in this case took the position that the claims on appeal here, claims 35 to 39 of
12 appellant's application were not patentable over the claims of the appellant's -- I beg your pardon, the plaintiff, we haven't reached that stage yet -- the plaintiff's patent No. 2,694,656.

THE COURT: Is that what is meant by double patenting?

MR. BEHRENS: Yes, Your Honor, this is one type of double patenting and the Board held with respect to this type of double patenting, it was well recognized in this circuit and in the Court of Customs and Patent Appeals that the claims of the second application, although co-pending with the patent already granted to the same inventor, must patentably and inventively distinguish from the claims already granted. It was the Board's decision here that the plaintiff's claims did not so distinguish and the Board used as an example claims 35 and 39 of the plaintiff's claims, and it held with respect to the claims of the case and particularly claims 35, 36, 37 and 38, which are directed to a process, that these claims failed to distinguish from claim 13 of the Camras patent when taken in view of certain publications which are given as Exhibits B, C and D of the Commissioner's exhibits.

Now, claim 13 of the patent is directed to a process of making a magnetic material in which you start with a non-magnetic ferric oxide or trivalent iron oxide material and treat it with hydrogen at certain temperatures, elevated temperatures; apparently there is no issue on that point, an elevated temperature is used for this part of the process
13 in both the patent claims and the application claims.

However, the plaintiff here is referring to a preliminary step. Taking claim 35 of the plaintiff's claims, the claim is emphasizing the preliminary step of heating the synthetic iron oxide to a temperature between 1000° and 1750° Fahrenheit. Now, looking at the claim 13 of the Camras patent, this is represented according to the Examiner by the step of providing -- let me read to be more specific here:

> "providing a synthetic non-magnetic ferric oxide in crystalline form and of a particle size not over 6 microns"

and it was the Board's position that the provision of this non-magnetic iron oxide, the starting material for this process of claim 13, is shown to have been accomplished in the prior art by the processes shown in the reference in "Nature," which is the Commissioner's Exhibit B, where a heating of iron oxide to a temperature of 700° is shown to produce a non-magnetic red oxide, and a process in Exhibit D of the prior art, Welo and Baudisch, and this reference shows that at a temperature of 550° C., the non-magnetic iron oxide is produced. And a temperature of 550° C. is equal to a temperature of 1022° F., which is right in the middle of the plaintiff's range of 1000° to 1750° F.

The Board took another approach on this. It held with reference
14 to these claims 35, 36, 37 and 38, that claim 21 showed the actual and claimed the actual pre-heat treatment that the appellants are stressing in this present suit. Looking at the Camras patent and looking at claim 21, the Board held that the pre-heating treatment between 500° and 1550° F. would, within the upper part of the range, result in this particular heat transformation to the non-magnetic iron oxide which is emphasized as an invention in the present case.

As to the product claim, 39 of the appellant, it was the Board's position that this claim was unpatentable over the product claims of the Camras patent, referring particularly to product claim 25 in Exhibit A. This product claim of the patent, claim 25, the Board pointed out was not directed to a cubic type of crystal, but the Board found no patentable distinction in this circumstance and they pointed to claim 4, which is directed to the product in its ultimate use, a magnetic impulse record member, that is the magnetic tape itself, in which no restriction as to a particle shape was found.

The Board pointed to the fact that various of these claims are not restricted to an acicular or needle-like form of iron oxide, so that no patentable distinction could be predicated on this in any event.

THE COURT: Do you have references to any Court of Appeals decisions on this special double patenting?

MR. BEHRENS: Yes, Your Honor, I believe I have. The most
15 recent of such decisions of the Court of Appeals was an unreported decision, Switzer v. Watson, appeal No. 13,948, which was decided on the last day of court and if I may pass it to Your Honor, the decision as sent to the Office, it is interesting because it affirmed a rejection on double patenting on a claim that is referred to here, which is exactly parallel to a case in the Court of Customs and Patent Appeals which was In Re Ward. It is an unusual parallel because the Judge in the District Court, Judge Holtzoff, made a specific finding on this question of double patenting. In Re Ward in the Court of Customs and Patent Appeals is reported at 236 F.2d 428, 43 CCPA 1007, and in this case an earlier patent was issued to Ward, a sole patent, and this dealt with the matter of discovering flaws in castings and they did it by putting a fluorescent material upon the surface of the casting and exposing it to ultraviolet light, I believe it was, and thereby spotting the flaw in the casting.

A second patent was requested in a joint application of Ward and Switzer, and in the Ward and Switzer application, instead of fluorescent material, a dye was used to spot the flaws in the castings and the claims in the patent, the application that first went to patent, did not cover the use of the dye. However, the Court of Customs and Patent Appeals and the District Court found that there was no patentable distinction between the claimed process with the fluorescent material in the patent and the
16 process in the application claims of using a dye material.

A previous reported decision of the Court of Appeals of this District Circuit on this matter is a case of Kistler v. Coe, which is 78 Appeals D.C. 36, 142 F.2d 94. The cases in the Court of Customs and Patent Appeals, as shown by In Re Mason, 20 CCPA 782, 62 F.2d 185, are based on a decision of the Court of Appeals of the District of Columbia, In Re Isherwood, 46 Appeals D.C. 507.

I believe the principle is well recognized, the difficulty of selecting the best cases --

THE COURT: It becomes a fact question, doesn't it?

MR. BEHRENS: I believe so, Your Honor.

MR. SHERMAN: May I just say, Your Honor, that the Patent Office has the advantage of us in that it has this unreported case of Switzer v. Watson. On the other hand, I am quite familiar with the case because we represent the Switzers in some other matters and there, as the Solicitor correctly pointed out but perhaps did not particularly emphasize, the sole and only distinction was between the use of a fluorescent dye in the issued patent and a non-fluorescent or what we call a visible dye in the second application. And here, in contrast, there is a new and novel step involving a distinctly different concept. I just want to point that out. We will have plenty of time to argue this, I don't doubt.

17 THE COURT: When he refers to an unreported decision, it merely hasn't been printed yet, it hasn't been put in the volumes yet; it will be in due course, I am sure.

MR. SHERMAN: Well, I will see it shortly.

THE COURT: I am sure I have one in my library.

MR. KRAUSE: Call Mr. Camras, please.

Thereupon --

MARVIN CAMRAS

called as a witness by the Plaintiff, being first duly sworn, was examined and testified as follows:

DIRECT EXAMINATION

BY MR. KRAUSE:

Q. Would you state your name, your address, and your age, please. A. Marvin Camras, 560 Lincoln Avenue, Glencoe, Illinois. Age, 42.

Q. What is your educational background? A. I have a Bachelor of Science degree in Electrical Engineering from Armour Institute of Technology, and a Master of Science degree from Illinois Institute of Technology, also in Electrical Engineering.

Q. What is your present position? A. I am a senior physicist at Armour Research Foundation.

Q. And how long have you been in that capacity? A. In the capacity of senior physicist about six or seven years, although I have been
18 at Armour Research Foundation since 1940.

Q. What is the business of the Armour Research Foundation? A. Their business is industrial research.

Q. Is the Armour Research Foundation the plaintiff named in this suit? A. Yes.

Q. How long has Armour Research Foundation been engaged in the activity of magnetic recording? A. Since 1940, about seventeen or eighteen years.

Q. Were you responsible for initiating that work there? A. Yes.

Q. What was the state of the magnetic recording art, so called, when Armour Research Foundation took an interest in it? A. I believe Mr. Sherman has summed up some of the early work of Mr. Poulsen in Copenhagen, Denmark, in the 1900's. There were a number of attempts in the 1930's in England and Germany to make a practical and commercial magnetic tape recorder but, as far as I know, there was no such machine until the war.

Q. How many United States patents are there in which you are named as inventor? A. About a hundred issued United States patents.

Q. Can you give me an estimate as to how many foreign patents
19 are in the same classification? A. I believe somewhere between a hundred and two hundred.

Q. These are all inventions in magnetic recording, then? A. Yes, mainly.

Q. Have you ever testified as a witness in relation to patent litigation concerning your patents? A. Yes, in interferences, and in some preliminaries, too, of litigation.

Q. Have you ever assisted in the preparation of patent applications directed to your inventions? A. Yes, I have.

Q. Have you ever received any other honors for your work in magnetic recording, beside the patents that you mentioned? A. I received a distinguished service alumni award of Illinois Institute of Technology in 1948, and a United States Camera award for contributions to sound on film in motion picture photography, and I received the John Scott Medal of the City of Philadelphia in 1955.

Q. Who are some of the other people who have received that John Scott Medal through the years? A. Thomas Edison, Madam Curie, Brittain and the other inventors of the transistor, Fleming of penicillin, and a number of other people that I can't think of.

Q. Has anyone in the magnetic recording field, other than yourself, been so honored? A. No, sir. 20

Q. What were the earliest types of magnetic recording media used that you can recall? A. The earliest types were solid materials, mainly wire but also tape, solid metallic materials.

Q. What were some of the drawbacks, if there were drawbacks, to this type of media? A. It had to be run at very high speeds and it was bulky and heavy and its sound quality was very poor, as compared to phonographs and other available methods for recording sounds, so that it was in no way competitive with these better known materials.

Q. What characteristics are required in a good magnetic record member? A. It must have magnetic properties which allow it to retain a good deal of magnetization as stored energy so that it can play back on subsequent runnings through the device.

Q. What is meant by, for example, coercive force and why is that important to magnetic record members? A. The coercive force is a measure of the ability of a magnetized material to retain its state of magnetization; in other words, the resistance to demagnetizing effects which would reduce that magnetization.

Q. Does coercive force also play a part in the spectrum or the fidelity of the sounds which you can record upon a given medium? 21

A. Yes, it does. We established that the coercive force determines the high frequency response of the record medium directly, and indirectly the speed at which it can be run. In other words, a high coercive material allows more information per inch to be recorded on a magnetic record material, so for the same sound quality the record material may be run slower if it has a high coercive force, or if it is run at the same speed it will have better sound quality than one which has a low coercive property.

Q. What is meant by the term "remanence" and how is that important in magnetic recording work? A. Remanence is the amount of magnetization retained by a record member and that would determine the loudness, especially at low and medium frequencies, that the sound would have.

Q. Are the properties required in the materials suitable for magnetic recording purposes different from the properties of other types of magnetic oxides? A. Yes, they are quite different. In general, we may classify magnetic materials as hard and soft. The soft magnetic materials are used in transformers and it is required of soft magnetic materials that they lose their magnetization very readily so that they do not become permanently magnetized. These are unsuitable for magnetic recording because they will not retain any magnetization. Hard magnetic
22 materials are used in permanent magnets such as in loudspeakers, in electric meters, and in similar instruments.

However, the commercial high coercive materials are too high for magnetic recording; if we try to use those, we find we cannot magnetize them to the proper extent with any possible or available or known magnetic recording head. Also, once magnetized to some extent, it is very difficult or practically impossible to erase such standard permanent magnetic materials. So a material which is to be suitable for magnetic recording must be a compromise, it must be a hard magnetic material but not too hard, so that it will retain its magnetization and yet be erasable so that it can be used over and over again.

Q. These magnetic materials about which you have been testifying, are they generally in a specific family of chemical compounds? A. Oh, there are almost an infinite number of possible magnetic materials but the ones which have been found valuable at least in our work, are magnetic iron oxides which have been given certain heat treatment.

Q. Are there several different types of oxides of iron? A. Yes, there are a large number of oxides of iron and there are at least four or five very definite and distinct chemical compounds, each one of which may have a very wide variety of magnetic properties ranging from completely non-magnetic to too highly magnetic for recording use.

23 Q. Well, is it possible then for two materials, each having identically the same chemical composition, to have very widely different magnetic properties? A. Yes.

Q. Would you please describe your earlier developments in the field of making magnetic record tapes? A. Well, in some of the very earliest work, I duplicated some of the prior art low coercivity materials. Actually, they thought they were high coercivity, which they were at the time.

THE COURT: High what?

THE WITNESS: I should say coercive force materials technically, we call it coercivity but I will try to use the words coercive force to be consistent. These materials had a coercive force ranging from 80 to about 120 and they were referred to in those days as high, and they were about as high as could be obtained in those days.

However, in our early patent, we found a certain type of heat treatment for a special material having an acicular or needle-like form, which allowed us to obtain coercive forces over 300 or, in other words, approximately three times as high as was possible with the earlier known materials. This enabled us to run the record material at one-third the speed with the same quality as was obtained before, and allows for an economy of material.

24 BY MR. KRAUSE:

Q. This patent you just referred to is No. 2,694,656, I believe, is that correct? A. Yes.

THE COURT: Is that a tape?

THE WITNESS: That is a powder which is mixed with a binder or glue and coated onto a paper or film backing, and the powder is the active material, but it consists of a thin layer on an inert carrier material.

BY MR. KRAUSE:

Q. Is the process and composition described in that patent, which we will refer to as the '656 patent, is that being used commercially now? A. Yes, it is. Minnesota Mining is one of the principal producers of magnetic tapes and they are making this under license from Armour Research Foundation.

Q. Do they sell under a brand name? A. Scotch brand.

Q. Approximately what percentage of magnetic tapes sold today are being manufactured under the process of your issued '656 patent, can you tell me? A. I would estimate that about 95% of all the tape which is used and sold is made according to our '656 patent.

Q. Mr. Camras, I am going to hand you a copy of that '656 patent, which has already been introduced by the Solicitor as an exhibit, and I call your attention to claim 13 of that patent. You will note that the first two lines read:

> "The method of making magnetic iron oxide which comprises providing a synthetic non-magnetic ferric oxide in crystalline form . . ."

Now, Mr. Camras, would you point out where, in the specification of your patent, there is a basis for that word "providing"? A. Yes, our patent gives Examples I to IX in columns 3 to 7, and these examples outline very definite materials which we recommend be used as starting materials.

Q. What is the crystalline shape of those materials? A. Those materials in general, and especially the one made in Example I, are acicular or needle-like in form. There is one exception in Example VIII, but in general they are acicular.

Q. Can you describe briefly the physical steps involved in making acicular type particles? A. Yes. The step consists of taking a salt of iron and precipitating it with an alkali and that produces a very finely divided ferric oxide which is used as a nucleus or seed. This seed is placed in a larger dilute solution of ferrous salt and allowed to grow, and in growing it forms a long needle-shape type of crystal. It is a yellow material and has the composition $Fe_2O_3.H_2O$. The H_2O is an integral part of the material and gives a yellow product. Without the H_2O, the product is not discolored and actually is of a different composition.

26 Q. Well, that procedure that you outlined there, is that the basis for the word "providing" in claim 13 of your patent? A. Yes, that is the preferred material which is to be used.

Q. Now, as these acicular particles are prepared, do they contain water which is part of the chemical structure of the oxide? A. It is closely associated with it. I am not a chemist, so I can't tell you the theory of whether it is really considered a part of it, but it is so closely associated that without the water of hydration, it loses its identity as that particular compound.

Q. Now, if you wanted to remove this water of hydration, how would you do it? A. It can be done by a heating step.

Q. At approximately what temperature? A. The water of hydration will start to come off at about 500° Fahrenheit and it takes a longer time to do it at this temperature, so that we may use higher temperatures and we specify in Example III, 500 to 1500° Fahrenheit as being suitable to drive off such water of hydration.

Q. You follow this dehydration treatment in every instance, how-
27 ever, with a subsequent treatment to improve the magnetic property, do you not? A. The hydrogen reduction treatment, which is used in various examples, is at a temperature high enough so that the water of hydration will be driven off at the time we are performing the heat treatment and hydrogen step, so that is automatically done whether we do it separately or not.

Q. Is there any advantage of doing it separately, outside the hydrogen furnace? A. There is the advantage of saving time and hydrogen material. It is a rather time consuming and hazardous operation, and the shorter the time cycle for reducing in hydrogen, the more batches that can be run in one day, so there is an advantage in driving off the water as a separate step and thereby shortening the cycle.

Q. I call your attention to claim 21 of this issued patent '656, specifically the portion thereof which reads:

> "heating said crystals in the air to a temperature between 500 and 1550° F. to drive off the water of hydration . . . "

Is that the type of step about which you have just been testifying?
A. Yes, it is.

Q. Are there other types of particles, aside from the acicular or needle type particles, which do not respond readily to the type of reduc-
28 tion treatment described in that patent? A. Yes, there are.

Q. How are these other types of particles prepared and what are these other types? A. Well, one type is a cubic type of oxide having the chemical composition Fe_3O_4. It is a black oxide and it is prepared by precipitation in a solution containing an oxidizing agent.

Q. Would you explain what you mean by oxidizing agent, Mr. Camras? A. Well, an oxidizing agent is one which has oxygen available and which yields such oxygen to the material which you are interested in reacting with.

Q. And you testified that cubic particles are made by this process? A. Yes, this process forms cubic particles directly and these have a different chemical composition from what we have talked about before. They are Fe_3O_4 instead of Fe_2O_3 and also they contain no water of hydration.

Q. So it is unnecessary, with that type of particle, to use a preliminary dehydration step, is that correct? A. That is right. The preliminary dehydration step would not do anything because it does not contain water of hydration.

29 Q. What can you say as to the magnetic properties of the cubic particles as they are originally prepared? A. The cubic material Fe_3O_4 as prepared is magnetic but has a low coercivity; in fact, it is the identical material which was used in one form of the prior art magnetic

tapes. Material as made by my description was mixed with a binder and used directly on the backing for a magnetic tape.

Q. Well, do the cubic particles that you have referred to respond in the same way the acicular particles do, to the subsequent hydrogen treatment? A. No, they do not. Treating them with hydrogen in accordance with the examples, does not enhance the magnetic properties appreciably.

Q. What additional techniques are necessary to bring the cubic particles up to the quality of your acicular particles? A. I found that a high temperature pre-treatment in air, which renders this cubic material non-magnetic, was essential in order to make it respond to the further treatment according to our '656 patent.

Q. Am I correct in saying, then, that the sequence is to take a cubic material which is already somewhat magnetic, demagnetize it, and then go back and magnetize it to the extent of producing a commercial record member? A. Yes, by that we refer to chemical treatments which render this low coercive material non-magnetic and later subjecting it to a series of steps involving reduction to render it magnetic again, and after the second treatment it becomes a high coercive material comparable to the acicular particles which are the main subject matter of the prior patent.

Q. This high temperature treatment about which you are testifying, is that the process described in the application which is in issue in this lawsuit? A. Yes, it is.

Q. Are you the applicant for patent in that application? A. Yes.

Q. Have you ever prepared samples to illustrate these various differences, about which you have been testifying, between the cubic and the acicular particles, for example? A. Yes, I have coated five samples of tape, three with cubic particles according to or as illustrated in various examples of our present application, and two with acicular particles made according to our prior patent.

Q. Mr. Camras, I hand you two charts entitled respectively,

"Relative Output of 2000 Cycle Signal" and "Relative Output of 4000
Cycle Signal" and ask you if you can identify these, please? A. Yes,
these charts are graphs of the sound level which came out of this ma-
31 chine when playing back the five different kinds of tapes.

THE COURT: You say "this machine," what machine?

THE WITNESS: I am referring to a little dictation type of tape recorder called the DeJur Grundig Stenorette.

MR. KRAUSE: I am asking that the two charts the witness has been referring to be marked as Plaintiff's Exhibits 2 and 3, the 2000 cycle chart being Exhibit 2 and the other being Exhibit 3.

THE COURT: They will be received.

(Chart of Relative Output of 2000 Cycle Signal was marked Plaintiff's Exhibit 2 and received in evidence.)

(Chart of Relative Output of 4000 Cycle Signal was marked Plaintiff's Exhibit No. 3 and received in evidence.)

BY MR. KRAUSE:

Q. Mr. Camras, would you explain the legend on the top of those
two charts? A. Well, the first tape is cubic particle material which
has been reduced at 700° in hydrogen, in accordance with our '656 patent.
The second one is the cubic material which has been given a pre-treat-
ment at 500° in air and then followed by a subsequent treatment at 700°
in hydrogen. This is to illustrate that a low temperature pre-treatment
in air still does not render the material suitable for forming a high
32 coercivity material. The third one --

Q. Excuse me. I think it would be easier if we could pass these up to His Honor and let you testify from copies of them. A. Shall I continue or start over?

THE COURT: You go right ahead.

THE WITNESS: The third one which is referred to as cubic was given a treatment of 1300° Fahrenheit in air followed by 700° in hydrogen, in accordance with the method taught in our present application and this had a high coercivity and gave a high output comparable to the acicular materials. The last two materials, I think the legends by now are self-explanatory, they are acicular materials made in accordance with the '656 patent and they show what happens when we use a preliminary step of 500° in air and a preliminary step of 1300° in air; and actually the treatment in air whether at high or low temperatures does not change the magnetic properties very appreciably and does not confer upon this material exceptional magnetic properties. This material does not require any of the treatment shown here in order to have the magnetic properties.

BY MR. KRAUSE:

Q. Then to summarize your testimony, you prepared five separate samples using in three instances cubic materials being treated under various conditions, and in two instances acicular materials for purposes of
33 comparison, is that correct? A. Yes.

Q. Did you assemble all these samples into a single tape? A. Yes, I spliced about ten-second lengths of each one of the five different types of tape, one right after the other.

THE COURT: Are these all on the '656 patent?

MR. KRAUSE: No, Your Honor. We are here drawing the distinction that, for example, in the first section of the Exhibit 2, we illustrate what happens when the cubic particles are merely given the treatment of the '656 patent without the improvement mentioned in this invention. The second section of that --

THE COURT: I understand.

BY MR. KRAUSE:

Q. How did you record the signal on the spliced tape that you mentioned? A. The signal was recorded by holding a tone generator or oscillator with a loudspeaker in front of the microphone and this was held perfectly constant and the level kept constant, so that all tapes were recorded under the same identical conditions.

Q. Well, with the indulgence of the Court, we should like to play that spliced tape sample.

THE COURT: All right. We usually take a short recess about this time, we will do that and then resume.

34 MR. KRAUSE: Thank you.

(A short recess was taken.)

BY MR. KRAUSE:

Q. Mr. Camras, before we go into a demonstration, would you summarize in your own words, briefly please, just what is shown on those two exhibits, 2 and 3, that you have before you? A. Yes. We show an oxide first, the first one is a cubic oxide which has been treated in accordance with our '656 patent and this shows that when we do that, follow the exact procedure of the '656 patent, the cubic oxide does not respond and does not yield a good high coercive material. In the second one, we have followed a procedure where we added an oxidation step but at a low temperature and we are showing that the temperature at which the oxidation step is carried on is also critical, because at 500°, although there is a chemical change and the Fe_3O_4 changes into Fe_2O_3, the subsequent treatment according to our '656 patent still does not render it into a high coercive material.

THE COURT: The results are almost the same.

THE WITNESS: Yes, it is almost the same; in fact, when we show the highest range of the magnetic properties, it is very slightly worse. However, when we give it the preliminary treatment, according to our present application, in air at high temperature, 1300°, then it results

35 in very high magnetic properties comparable to the magnetic properties which acicular particles obtain when given the treatment in our patent without requiring the preliminary high temperature treatment.

BY MR. KRAUSE:

Q. That is reflected in section three of that graph there? A. Yes, in section three of the graph, we show the good cubic material which has been obtained as a result of treatment according to our present application.

Q. Would you go on to section four? A. Section four and section five are the acicular materials which are made in accordance with our '656 patent and which have the additional aeration step described in claim 21, and it shows here that the steps of either 500 or 1300° do not produce any appreciable difference. The material is good regardless of whether you use these high temperature pretreatments or, as indicated in our '656 patent, whether you do not use it at all, because the material in our '656 patent, other than in claim 21, is not made with any preliminary high temperature treatment and with the acicular material, they are all good.

Q. I believe you testified earlier that you had coated some tapes. Now, just to give us a physical idea of what you did, you have samples of tapes which have been coated? A. Yes. These are runs which I
36 made at the same time. In fact, they are runs during the time I was making tapes that we are demonstrating on the machine and this is the way it comes out of our coating machine. You note it has cellulose acetate backing and on it is painted a strip of oxide material. The material is then slit or sliced into quarter-inch wide tapes.

Here is a sample of what one of these is like, these are just slit out of the portion which is coated and they fit on these tape recorders. Here is a tape recorder and we show the tape is wound on reels and is run back and forth against a magnetic head which magnetizes the tape in accordance with the sound impressions.

Q. This machine you have here, is that a commercially available magnetic recorder? A. Yes, this is a commercially available magnetic recorder which is widely used for dictation purposes.

Q. Would you explain in advance, please, what we will hear when we hear this tape sample played? A. Well, you will hear these five samples in succession, each one taking about ten seconds. The first two will be the cubic materials which have not been treated adequately, the third one will be the cubic material which has been treated according to the present application, and the fourth and fifth are acicular materials which are conventionally used in tapes, but which have been made spe-
37 cially for this experiment, keeping the thickness and the proportion and everything else absolutely identical for all five tapes. The only variation is in the treatment of the powders.

MR. KRAUSE: May we play this sample?

THE COURT: Yes.

THE WITNESS: This is the first tape, cubic at 700°. Now, we are on the second one, you will notice there is hardly any difference in the sound level. This is the third or improved material. Now, we are on the acicular material which has been given the 500° aeration treatment. And finally, we are on the acicular material which has been treated at 1300° in air.

We notice the last three are approximately the same level, so that the improved cubic material compares with the acicular. (Playing of tape concluded.)

BY MR. KRAUSE:

Q. Did you measure the magnetic particles of the cubic particles which were not given the high temperature treatment? In other words, portion one of the chart there? A. Yes, I did.

Q. What result did you obtain? A. I believe I have that somewhere else. It is in a folder on the desk there. (Folder handed to witness.)

The cubic material reduced at 700° in hydrogen, coercive force,
38 160.

Q. Would this type of material be suitable for magnetic record members as used today? A. No, it would not.

Q. Are you familiar with the article written by Welo and Baudisch entitled, "The Two-Stage Transformation of Magnetite into Hematite" which is of record in this proceeding? A. Yes, I am.

Q. I hand you a copy of that. Specifically, what does this article disclose? A. This article discloses the chemical changes which take place when an iron oxide is heated at various temperatures.

Q. Does it have anything to do with magnetic recording? A. Nothing at all.

Q. Is it primarily a chemical treatise or magnetic treatise? A. It is primarily a chemical treatise. I think they mention that the material has magnetic properties but it doesn't say what they are, and actually the low coercivity properties are the ones which are referred to, from my experience with such material.

Q. Does that article have to do with the preparation or synthesis
of oxide materials or merely the treatment of materials which have
previously been prepared? A. It has mainly to do with treating a ma-
39 terial which has been previously prepared.

Q. Does this Welo and Baudisch article disclose any composition which can be used as is for magnetic recording purposes? A. Not for a modern record member, although I think I mentioned before that such material was actually used in prior art recording tapes.

Q. Are you familiar with the article appearing in Mellor, "Inorganic and Theoretical Chemistry" which was cited as a reference against your application? I hand you a copy of the pertinent pages thereof. A. Yes.

Q. Can you tell us briefly what that article discloses? A. This article is concerned with chemical reactions that take place in magnetic oxide when given various treatments.

Q. Does that article have anything to do with magnetic recording? A. No, it does not. It is in a treatise on "Inorganic and Theoretical Chemistry," it says on top of the page.

Q. Is there any composition disclosed therein which would have applicability as is to modern day magnetic recording? A. No, there is not.

Q. Are you familiar with the article appearing in "Nature,"
40 authored by Abraham, which is a reference against the claims of
your application? I hand you a copy of that. A. Yes, I am.

Q. Can you describe briefly what is shown in that reference, please? A. This tells the chemical changes which take place when iron oxide is given various treatments. It also mentions that there are magnetic properties, but does not say what they are.

Q. Does that article disclose any composition which is useful as is in modern day magnetic recording? A. No, it does not.

Q. Summing up, Mr. Camras, what did you find in your work was the difference in behavior between the acicular particles and the generally cubic or equant particles, toward the acquisition of the desired magnetic particles by the step or steps of reduction followed by oxidation? A. Well, the steps of reduction or reduction followed by oxidation, would directly and without anything further give a high coercive material suitable for magnetic recording, and this is what is disclosed in our '656 patent.

Q. Now, you are referring to the acicular particles, are you not? A. I am referring to the acicular particles.

Q. That was not true of the cubic particles, is that correct?

41 A. That was not true of the cubic particles.

Q. What solution did you provide to make the cubic particles the equivalent of the acicular particles mentioned in your patent? A. I found that a high temperature treatment prior to following the steps of our '656 patent gave an entirely different result when the cubic particles were used and allowed these cubic particles, after such treatment, to have the desirable magnetic properties.

Q. Do you find your solution to the problem suggested in any of the claims of your issued '656 patent? A. The only one which could possibly have anything to do with it would be claim 21, but that has to do with dehydrating acicular crystals and, as I have indicated and as we have shown on the tests, the dehydration step, regardless of what temperature it is carried on, does not confer any specially desirable magnetic properties on the acicular material nor is it required in the case of the acicular material.

Q. And I think you testified also that the cubic particles do not have water of hydration, so it would not be necessary to use that step, is that correct? A. No, this could not apply to the cubic particles because they do not have any water of hydration; therefore, they can't be dehydrated.

42 Q. Do you find any suggestion of your new improvement in the prior art cited against your application in the Patent Office? That is, the combination of the steps of taking an already magnetic oxide,

intentionally demagnetizing by high temperature treatment, followed by reduction, to produce an acceptable record method? A. No, I don't.

Q. Would you expect to find a solution of this type in the type of art which the Patent Office has relied upon? A. No, I would not expect to find a solution of that type in this art, because this art is of, oh, you might say, in the realm of pure science where they are interested in reactions and what takes place; in other words, the nature of the materials themselves and without any study or without any regard as to their suitability for any application.

Q. I call your attention, Mr. Camras, again to claim 13 of your '656 patent, which contains the phrase: "providing a synthetic non-magnetic ferric oxide . . ." Do you find any correlation between the words "providing" and "synthetic"? A. Well, we have disclosed in our Examples I to IX various chemical reactions for synthesizing this oxide starting from chemicals, you might say. In other words, you can buy some ferrous sulphate crystals and, starting with these and similar chemicals, synthesize an iron oxide; in other words, make it from its elements.

43 Q. Would you consider heating an existent iron oxide above the Curie point, a synthesis of the oxide? A. No, I would say that is a transformation but it is not a synthesis. You have not synthesized it from prior elements, from similar elements.

MR. KRAUSE: No more examination, Your Honor.

CROSS-EXAMINATION

BY MR. BEHRENS:

Q. Mr. Camras, excuse me, is it Dr. Camras? A. No, Mister.

Q. Would you take Exhibit 2 for the Plaintiff, 2 or 3, it doesn't make any difference. A. Here is 3.

Q. Looking at Exhibit 3 and looking particularly at the first column there, "Cubic - 700H," does that material respond to the description of the product of claim 13 of the Camras patent?

MR. SHERMAN: Claim 13 is a method claim and you are talking about a chart.

MR. BEHRENS: I say, respond to the product.

BY MR. BEHRENS:

Q. Could I hand you, to help you out in that matter, the Defendant's Exhibit 1, Item A, the Camras patent. A. Yes, I have it here.

Q. Would you look at claim 13 and tell me whether the product
44 referred to in column 1 of your chart, Exhibit 3, corresponds to
the product, the description of that product in the claim? A. Well,
for one thing, it calls for a ferric oxide and this cubic material is what
is known as a ferrosoferic oxide, so in that respect the claim would not
apply to that.

Q. I refer you to: "produce a ferrosoferric oxide having permanent magnet properties including a coercive force of at least 200 oersteds." A. Well, in that respect it also does not respond, because this material I believe I said to have a coercive force of 160 oersteds, which is well below 200.

Q. In other words, that would not be the product of claim 13? A. No, it would not.

Q. Nor would the product used in the second column refer to the patented product produced by the process of claim 13? A. No, the product would not, but in this case we have a ferric oxide that is referred to in the second line of claim 13, so we are starting and applying the treatment to a ferric oxide.

Q. But in both cases you get a product in both columns one and
two of both graphs which does not have the 200 oersteds required by the
patent claim, is that right? A. That is correct. In fact, I have a
45 notation here that the second one had a coercive force of 140.

Q. In treating this cubic iron oxide which is referred to in column one of your Exhibits 2 and 3, was the original iron oxide magnetic or non-magnetic? In other words, was the iron oxide which was treated at 700° Fahrenheit in the first column, before it was treated, was it magnetic or non-magnetic? A. It was magnetic.

Q. It was magnetic? A. Yes.

Q. In other words, the product that was used in these tests failed to meet one of the other requirements of claim 13 in that it was -- I beg your pardon, I am talking too much.

MR. SHERMAN: I'd like to have the witness answer that, I think it is a good question.

THE COURT: Had he finished his question? I think he stopped half-way.

MR. SHERMAN: I didn't hear Your Honor.

THE COURT: I think he had not finished his question. He decided to withdraw it.

MR. SHERMAN: We will ask it.

THE COURT: Did you withdraw your question?

MR. BEHRENS: I will ask it. I think it has already been stated.

BY MR. BEHRENS:

46 Q. Were the same type of materials used in, say in the patent claim process and in the process producing the product used in the first column? A. Can you explain further what you mean by used in the patent process?

THE COURT: You mean in claim 13?

BY MR. BEHRENS:

Q. In claim 13, non-magnetic iron oxide is referred to as the initial reactant. Did you use a non-magnetic or a magnetic iron oxide to prepare your material for column one? A. This cubic material is inherently magnetic.

Q. It was magnetic? A. It was magnetic.

Q. Looking at the disclosures of Welo and Baudisch, would any treatment shown in the Welo and Baudisch article produce a non-magnetic iron oxide which would be useful as a starting material in your process, the process of the application? A. I am not sure, because Welo and Baudisch don't give any details of how to synthesize the material, so I am not certain whether this material, regardless of the various stages that we take it at, whether at any stage it would yield a high coercive material. In other words, I have had enough experience to know that I can't determine in advance whether it will or not, from the information that is given.

47 Q. I assumed that any non-magnetic iron oxide of the proper particle size, of course, would yield to your treatment of high temperature preliminary treatment and a later hydrogenation to yield a product that can be used with the properties that you call for in your claims? A. No, that is not true. There are some materials which will not respond.

Q. You referred, I believe, to Mellor as being a theoretical chemical type of publication and directed more nearly to chemistry and the theory of chemistry than a magnetic reproduction. Are not the claims of this case directed to a chemical treatment? A. Only partly, because I don't think there is any claim here which does not recite the magnetic properties that are required. In other words, the chemical treatment is a means toward an end in these claims.

Q. In every case we have a chemical treatment, I understand. Would you not term hydrogenation or reduction a chemical treatment? A. Yes.

Q. How can you distinguish between a synthesis and a treatment
on the basis of the materials treated? A. As I understand it, synthesis
is the formation of an entirely new material from its basic components;
in other words, putting together various elements into a more compli-
cated compound, whereas a treatment might make no change whatever
48 in the chemical composition and that is true of many of the in-
stances here, or it may involve simply adding another proportion of an
element, such as oxygen, which is already present; in other words, chang-
ing it to a higher degree of oxidation, perhaps, or reducing it to a
lower degree of oxidation, but having essentially the same type of com-
pound.

Q. What is the starting material in your synthesis to produce the magnetic material that you claim in your application? A. Well, the basic starting material that we show is ferrous sulphate, $FeSO_4 \cdot 7H_2O$.

MR. SHERMAN: I think the witness didn't quite understand the question, because the question referred to the application, meaning the application in this suit, and I believe you are looking at the patent.

THE WITNESS: Oh, I am sorry.

THE COURT: Do you now understand the question?

THE WITNESS: I do, but I don't have a copy of the application.

MR. BEHRENS: Could we hand the witness a copy of the application? Perhaps it would be easier to refer you to the Defendant's Exhibit 1, Item F. Do we have that here?

BY MR. BEHRENS:

Q. Looking at claim 35 on the first page of that, the decision of the
49 Board, but they include a copy of the claim, what is the starting material on the treatment there? A. It says "from a starting synthetic iron oxide containing a trivalent iron atom . . ."

Q. Do you regard changes in that iron oxide as being a matter of treatment or synthesis? A. I would say they are a treatment.

Q. Looking at the patent which is the Camras patent, which appears as Item A in the Commissioner's Exhibit 1, and looking at column 2 which is on page one, and starting at line 50, would you say that that restricts your --

MR. BEHRENS: I beg your pardon, Your Honor. The thought occurs to me that I am asking questions about something that is on the record and I would be wasting the Court's time. I withdraw the question.

THE COURT: Very well.

MR. BEHRENS: That is all, Your Honor.

THE COURT: Do you have any redirect?

MR. KRAUSE: No more examination, Your Honor. Thank you.

(Witness excused.)

MR. SHERMAN: What is your pleasure as to briefs, Your Honor?

THE COURT: What do you want to do?

MR. SHERMAN: Well, thirty and twenty days? Thirty, twenty and ten?

THE COURT: Make it twenty and ten. Do you have anything
50 else then?

MR. KRAUSE: No, Your Honor.

THE COURT: Thank you very much. I will await your briefs.

(Whereupon, the foregoing proceedings were concluded at 12 o'clock noon.)

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58 [Filed Mar. 31, 1958]

MEMORANDUM

This is an action brought under Title 35 U.S.C.A., Section 145 to authorize the Commissioner of Patents to issue to the plaintiff Letters Patent containing Claims 35, 36, 37, 38 and 39 of application Serial No. 114,677, entitled "Improved Coating Material For Magnetic Recording", and filed by Marvin Camras, assignor to the plaintiff herein, on September 8, 1949. Claims 35 through 39 are as follows:

> 35. In a method of making permanent magnet material from a starting synthetic iron oxide containing a trivalent iron atom having a coercive force less than 200 oersteds and having a particle size of one micron and less by reducing said iron oxide at an elevated temperature, the improvement which comprises the preliminary step of heating such starting synthetic oxide to a temperature between 1000° and 1750° F. for a sufficient length of time that when subsequently reduced at an elevated temperature below that used in said preliminary heating step said iron oxide acquires a coercive force value in excess of 200 oersteds and higher than could be obtained by employing the same reducing conditions in the absence of said preliminary heating step.
>
> 36. The method defined by claim 35 in which the preliminary heating step is carried on in the presence of an oxidizing atmosphere, and the subsequent reduction is carried out in a reducing atmosphere at a temperature of between 400° and 1200° F.
>
> 59 37. The method defined by claim 35 in which the starting synthetic oxide is of cubic particle shape, the preliminary heating step is carried out in the presence of air, the subsequent

reduction step is carried out in a hydrogen atmosphere and the resulting reduced iron oxide has a coercive force value in excess of 300 oersteds.

38. The method defined by claim 35 in which the starting synthetic oxide is ferrosoferric oxide (Fe_3O_4) of cubic particle shape, the preliminary heating step is carried out in the presence of air, the reduction step is carried out in a reducing atmosphere and the resulting reduced iron oxide is re-oxidized at a temperature from about 400° to 900° F. in an oxidizing atmosphere to produce an iron oxide having a coercive force value in excess of 200 oersteds.

39. Permanent magnet material adapted to form an element of a magnetic impulse record member consisting of synthetic magnetic iron oxide particles of a cubic particle shape less than one micron in size and having a coercive force value (H_c) between 200 and 450 and a remanence (B_r) of 200 and above.

The Patent Office Board of Appeals, in a decision rendered on March 8, 1956, affirmed the decision of the Primary Examiner with respect to the above quoted claims; finding them unpatentable because they were contrary to the law against double patenting. Sections 13, 21 and 25 of United States Patent No. 2,694,656, issued to Marvin Camras on November 16, 1954, along with secondary references, were used to support the Patent Office decision.

Plaintiff has attempted to distinguish the pending application from the issued patent by the use of a high temperature pre-treatment which results in higher coercive force values for materials which, without the pre-treatment, would not be effective under the issued patent. The Patent Office says that materials to be used in the issued patent are not limited to exclude the materials to be used under the pending application. Moreover, the issued patent provides for a step of heating the iron oxide crystals to a temperature between 500° and 1550° F., which step corresponds to the plaintiff's pre-heating step. The Patent Office further held that a conventional procedure used to reach the starting

material of the issued patent is by heating the iron oxide within the
60 range specified by the plaintiff's claim.

It is settled law that a patent should not issue for an invention which is covered by an issued patent, particularly to the same patentee, unless the matter described in the second application is essentially distinct and separable from the invention covered by the issued patent. Miller vs. Eagle Mfg. Co., 151 U.S. 186. As was held in In re Horneman, 194 F.2d 108, an inventor after obtaining a patent for a certain invention "may not, in a subsequent application, be allowed a patent containing claims which do not distinguish patentably over those of the issued patent." Also see Application of Greenlee, 222 F.2d 739.

In this case the claims of the plaintiff's application do not distinguish themselves patentably from those of the issued patent. Accordingly the complaint will be dismissed.

Counsel for defendant will submit proposed Findings of Fact, Conclusions of Law and Judgment.

/s/ Joseph C. McGarraghy
JUDGE

March 31, 1958

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66 [Filed April 30, 1958]

JUDGMENT

This action came on to be heard at last term and thereupon upon consideration thereof, it is this 30th day of April, 1958

ADJUDGED that the complaint be and it is hereby dismissed, with costs against plaintiff.

/s/ Joseph C. McGarraghy
JUDGE

Approved as to Form:
/s/ Smith, Michael & Gardiner
Attorney for Plaintiff

61 [Filed April 30, 1958]

FINDINGS OF FACT

1. Armour Research Foundation of Illinois Institute of Technology, the assignee of Marvin Camras, brought this action under 35 U.S.C. 145 to authorize the Commissioner of Patents to issue letters patent to it containing claims 35, 36, 37, 38, and 39, of the Marvin Camras application for patent entitled "Improved Coating Material For Magnetic Recording," Serial No. 114,677, filed September 8, 1949.

2. The Camras application relates to a process for preparing, by high temperature reduction, a finely divided magnetic iron oxide which is intended for use as the magnetic coating material on magnetic sound recorder tapes, and to finely divided, synthetic magnetic iron oxide particles having, as set forth in application claim 39, a cubic shape and a coercive force value "between 200 and 450" and a remanence "of 200 and above."

62 3. The emphasized feature of the process claimed in claims 35, 36, 37, and 38 of the Camras application is a preliminary step of heating a synthetic iron oxide having a trivalent iron atom and a coercive force less that 200 oersteds to a temperature between 1000° and 1750° F., so that the iron oxide, if magnetic, will permanently lose its magnetic properties, or will remain feebly magnetic. The subsequent reduction of this non-magnetic, or feebly magnetic, iron oxide at an elevated temperature below that used in the preliminary heating step is said to result in an iron oxide which has "a coercive force value in excess of 200 oersteds and higher than could be obtained by employing the same reducing conditions in the absence of said preliminary heating step." The process is described in the specification as applicable to, but not restricted to, iron oxide particles of a cubic shape, and claims 37 and 38 refer to the treatment of iron oxide particles of this shape.

4. Marvin Camras patent No. 2,694,656, which was granted on November 16, 1954, to the plaintiff as assignee of Marvin Camras, describes and claims a process of preparing magnetic iron oxides by the high temperature reduction of finely divided, non-magnetic ferric

oxide particles. Claim 13 of the Camras patent includes the preliminary step of "providing a synthetic non-magnetic ferric oxide in crystalline form and of a particle size not over 6 microns in its greatest dimension." Camras patent claim 21 recites the preliminary steps of precipitating crystals of $Fe_2O_3.H_2O$ (ferric oxide monohydrate) from solution and "heating said crystals in the air to a temperature between
63 500 and 1550° F., to drive off the water of hydration." Camras patent claim 25 is directed to particles of magnetic ferrosoferic oxide or ferric oxide in the form of elongated crystals less than 1.5 microns in maximum dimension, a coercive force between 245 and 330, and a remanence of above about 500 gauss. Although Camras patent claim 25 specifies that the iron oxide particles be elongated, other claims, including claims 4 and 13, are not so restricted and this absence of specific restrictions as to particle shape is consistent with statements in the specification of the patent.

5. The publications: Abraham *et al.*, *Nature*, Vol. 115, page 930, dated June 13, 1925; Welo and Baudisch, *Philosophical Magazine*, Vol. 50, pages 398-408, dated 1925; and Mellor, *Treatise on Inorganic and Theoretical Chemistry*, Vol. 13, pages 732, 736, 738, and 780, dated 1934, demonstrate that it has long been well known that the oxidation of ferrosoferic oxide, or the mere heating of ferric oxide, at temperatures above 550° C. (1022°F.) results in the production of a non-magnetic ferric oxide.

6. The provision of a non-magnetic ferric oxide, described as the first step of the process in Camras patent claim 13, by heating a starting synthetic iron oxide containing a trivalent iron atom to a temperature between 1000° to 1750° F., would not involve patentable invention in view of the noted disclosures of Welo and Baudisch, Mellor, and Abraham *et al.*

7. Process claims 35, 36, 37, and 38 of the Camras application
64 Serial No. 114,677 do not distinguish patentably from claim 13 of the Camras patent No. 2,694,656.

8. The preliminary heat treatment at temperatures between

1000° and 1750° F., the feature of Camras application claims 35, 36, 37, and 38 on which plaintiff predicates patentability, provides no patentable distinction from the heating of ferric oxide monohydrate at temperatures between 500° and 1550° F. which is called for in Camras patent claim 21.

9. Camras application claims 35, 36, 37, and 38 do not distinguish patentably from claim 21 of the Camras patent.

10. The oxide particles with a cubic shape called for in Camras application claim 39 are not shown to have magnetic properties which substantially distinguish them from the elongated iron oxide particles of Camras patent claim 25 or from iron oxide particles, without limitation as to shape, which have been prepared by the process of Camras patent claim 13 or utilized in the product of Camras patent claim 4.

11. Claim 39 of the Camras application defines no patentable invention over Camras patent claim 25, particularly in the light of Camras patent claims 4 and 13.

CONCLUSIONS OF LAW

1. An inventor after obtaining a patent for a certain invention may not, in a subsequent application, be allowed a patent containing claims which do not distinguish patentably over those of the issued patent.

65 2. The plaintiff is not entitled to a patent containing any of the claims 35, 36, 37, 38, or 39 of the Camras application Serial No. 114, 677.

3. The complaint should be dismissed as to all the claims in suit.

/s/ Joseph C. McGarraghy
JUDGE

April 30, 1958.

67 [Filed May 22, 1958]

NOTICE OF APPEAL, UNDER RULE 73, TO THE UNITED STATES COURT OF APPEALS FOR THE DISTRICT OF COLUMBIA

Notice is hereby given that Armour Research Foundation of Illinois Institute of Technology, (a corporation of Illinois), the plaintiff in the above-identified Action, hereby appeals to the United States Court of Appeals for the District of Columbia from the order dismissing the Complaint herein entered in this action on the 30th day of April, 1958.

/s/ Smith, Michael & Gardiner

* * *

/s/ Benjamin H. Sherman

/s/ O. R. Krause

Hill, Sherman, Meroni, Gross &
Simpson

* * *

Chicago, Illinois

Attorneys for Appellant,
Armour Research Foundation of
Illinois Institute of Technology.

Copy to C. W. Moore,
Solicitor, U. S. Patent Office

Date:

May 22, 1958.

68 [Filed June 4, 1958]

DESIGNATION OF CONTENTS OF RECORD ON APPEAL

Pursuant to Rule 75(a) of the Federal Rules of Civil Procedure, the Plaintiff-Appellant, Armour Research Foundation of Illinois Institute of Technology, hereby designates for inclusion in the record on appeal to the United States Court of Appeals for the District of Columbia, taken by Notice of Appeal filed May 22, 1958, the following portions of the record, proceedings and evidence in this action:-

(1) The Complaint for Issuance of Patent filed May 17, 1956.

(2) The Answer to the Complaint filed July 2, 1956.

(3) Transcript of Proceedings in the United States District Court for the District of Columbia.

69 (4) Memorandum decision of the Honorable Joseph C. McGarraghy, Judge, dated March 31, 1958.

(5) Judgment dated April 30, 1958.

(6) Findings of Fact and Conclusions of Law dated April 30, 1958.

(7) Notice of Appeal filed May 22, 1958.

(8) Certified copy of file wrapper and contents of the Marvin Camras Patent Application Serial No. 114,677, entitled "Improved Coating Material for Magnetic Recording", Plaintiff's Exhibit 1.

(9) References cited during prosecution of Camras Patent Application Serial No. 114,677 in the United States Patent Office, included in Defendant's Exhibit 1.

(10) Chart of Relative Output at 2000 Cycles, Plaintiff's Exhibit 2.

(11) Chart of Relative Output at 4000 Cycles, Plaintiff's Exhibit 3.

70 (12) This Designation of Content of Record on Appeal.

(13) Docket entries.

/s/ William A. Smith, Jr.

Smith, Michael and Gardiner

* * *

/s/ Benjamin H. Sherman

Hill, Sherman, Meroni, Gross &
Simpson

* * *

Chicago, Illinois

Attorneys for Appellant,
Armour Research Foundation of
Illinois Institute of Technology

[Certificate of Service]

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73 [Filed June 27, 1958]

ORDER TO TRANSMIT ORIGINAL EXHIBITS

Upon oral motion by counsel for Plaintiff and for good cause shown, it is by the Court this 27th day of June 1958

ORDERED: That the Clerk be, and hereby is, authorized to transmit original exhibits in this proceeding to the United States Court of Appeals for the District of Columbia Circuit for use in connection with the appeal herein:

(1) Certified copy of file wrapper and contents of the Marvin Camras patent application Serial No. 114,677, entitled "Improved Coating Material for Magnetic Recording", Plaintiff's Exhibit No. 1.

(2) Copies of all references cited during prosecution of Marvin Camras patent application Serial No. 114,677 in the United States Patent Office and included in Defendant's Exhibit No. 1.

(3) Chart of Relative Output at 2,000 cycles, Plaintiff's Exhibit No. 2.

74 (4) Chart of Relative Output at 4,000 cycles, Plaintiff's Exhibit No. 3.

/3/ Joseph C. McGarraghy
JUDGE

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75 [Filed Feb. 4, 1958, Harry M. Hull, Clerk]

PLAINTIFF'S EXHIBIT NO. 1
(Paper No. 1)

[Received, U. S. Patent Office, September 8, 1949]

114677

SPECIFICATION

TO ALL WHOM IT MAY CONCERN:

BE IT KNOWN that I, MARVIN CAMRAS, a citizen of the United States, and a resident of the City of Chicago, in the County of Cook, and State of Illinois, have invented certain new and useful improvements in

"IMPROVED COATING MATERIAL FOR MAGNETIC RECORDING"

and I do hereby declare that the following is a full, clear and exact description of the same.

The present invention relates to a method for producing an improved ferromagnetic material for magnetic recording purposes, and the product produced thereby.

More particularly, the invention relates to the preparation of ferromagnetic material for use in magnetic impulse record members comprising a base or carrier, such as a tape, ribbon or the like of non-magnetic material coated or impregnated with a track of ferromagnetic material.

Magnetic sound recorder tapes, as commonly produced, comprise a non-magnetic backing, such as paper, plastic, or the like, coated or impregnated with a magnetic oxide of iron. The ordinary magnetic oxides of iron heretofore used have not been entirely satisfactory since

76 such magnetic oxides do not possess magnetic properties particularly suited for magnetic recording purposes. For one thing, the more common ferromagnetic oxides of iron have relatively low coercive force values (H_c) and relatively low energy products. In addition, the ratio of the ferric induction (B_{fm}) to the residual magnetization (B_r) of the ordinary magnetic oxides is considerably higher than that required for best operation. The H_o value (the H value of the material at the point where the B_r versus H curve begins to rise rapidly) for such material is also low.

For magnetic recording purposes, an H_c above 200 and preferably from 220 to 450 is desired. The B_r of the coating should be 200 to 1000 or more, although high coercive force powders with lower B_r values may be used to advantage. The radio of B_{fm} to B_r should be low. Also, the B_r v. H curve should have very little B_r below the H_o point and rise rapidly and fairly linearly above the H_o point. A sharp saturation value is desirable, from the standpoint of erasing, with saturation preferably occuring between the field of 700 to 2000 oersteds, the lower values being preferred.

In my previous application Serial No. 763,486, filed July 25, 1947 and entitled "Permanent Magnet Material and Method of Making the Same", of which the present application is a continuation-in-part, I have described a particularly suitable magnetic iron oxide composition consist-
77 ing of particles having a maximum dimension of less than 6 microns and having an acicular crystalline form. It has been found that this particular crystalline form contributes much to the recording characteristics of the material. However, a new technique is necessary to impart adequate magnetic properties to particles as small as 1 micron or smaller in maximum dimension, regardless of the form of the individual particles.

In some applications, magnetic iron oxide particles are preferably in the form of cubes. Previous methods for enhancing the magnetic properties of cubic particles have not attained any great measure of success. However, I have found that the technique of the present invention is suitable for use in preparing coating materials from such cubic particles, especially where the size of the cubic particle is less than one micron in maximum dimension.

In still other applications, it would be highly desirable to use relatively inexpensive iron oxides whose particle sizes are relatively large and whose particles assume a great variety of indiscriminate shapes. The process of the present invention has also been found to enhance the desirable magnetic properties of such iron oxide particles above those obtainable by the use of prior methods.

78 An object of the present invention, therefore, is to provide a method for producing a coating material suitable for use in the coating or impregnation of magnetic strip material to form an improved magnetic impulse record member.

A further object of the present invention is to provide a method of enhancing the magnetic properties of extremely fine iron oxide particles which can be coated onto or impregnated into a record member for use in producing accurate recordings at slow speeds.

A further object of the present invention is to provide a method for enhancing the magnetic properties of powdered iron oxide, regardless of the shape of the individual particles, while still retaining their desirable physical characteristics.

Still another object of the present invention is to provide a method for producing magnetic particles having a high coercive force from relatively inexpensive iron oxide powder.

Magnetic oxides prepared by ordinary procedures usually have an Hc below 150, a B_r usually less than 400, a low H_o value, and a high ratio of ferric induction to residual magnetization.

I have now found that the magnetic properties of the various iron oxides, containing a trivalent iron atom, regardless of their physical
79 form, can be improved by means of a combination of heating and reduction steps. The process of the present invention is particularly applicable to the treatment of ferric oxide, Fe_2O_3, its hydrate, $Fe_2O_3 \cdot H_2O$, or to ferrosoferic oxide, Fe_3O_4.

In the first of these steps, the material is heated preferably in the presence of air or other oxidizing gases to a temperature in the range from about 1000 to 1750° F., and normally between 1100° F. and 1500° F. Higher temperatures may be used with favorable results for magnetic properties, although there is the danger that the particles may agglomerate or otherwise lose their desirable physical form. The resulting product is then reduced in a reducing atmosphere at a temperature below that employed in the initial heating stage. In general, the first heating step is carried out at or above a critical temperature where the oxide, if magnetic, will permanently lose its magnetic properties, or will remain just feebly magnetic.

Since the reactivity of iron oxides varies so greatly, no particular treating time can be given for all types of ferric oxides, nor is the time especially critical. For example, synthetically prepared ferric oxides are in general far more reactive than naturally occurring ferric oxides, and the heating time for such synthetic oxides will be very substantially less than that required for the treatment of naturally occurring oxides. In treating such reactive oxides, the desired change occurs almost instantaneously as soon as the proper temperature is reached. In many cases, the proper treatment time can be observed by observing the color change occurring in the material undergoing heat treatment. Ferrosoferric oxides which are normally magnetic will change from a black color to one having a reddish cast after a few minutes time in an oxidizing atmosphere. No noticeable harm occurs if the particles are kept at the temperatures given for prolonged periods of time, except that at the higher temperatures the oxide particles may tend to agglomerate.

The succeeding reduction step is carried out in a reducing atmosphere such as hydrogen gas or other reducing gas such as natural gas, carbon monoxide, city gas, and the like. Instead of using a reducing atmosphere, the reduction can be carried out at elevated temperatures in the presence of reducing agents such as pyrogallol, sulfur, or sodium acetate, as disclosed in my parent application Serial No. 763,486.

Here again the optimum temperature and time of treatment depends upon the reactivity of the individual particles. A synthetic, finely divided iron oxide powder will be completely converted into a good magnetic powder at a low temperature, whereas a less reactive powder will be unaffected by such a low temperature. In addition, the purity and dryness of the hydrogen also affects the length of the time and temperature to be used. While it is normally desirable to use pure hydrogen in the step, a small amount of water vapor may advantageously be added in order to prevent overbaking of the material, especially when the operation is carried out at higher temperatures. In general, however, a temperature from 400° to 1200° F. and preferably from 500° to 800° F. has been found best suited for carrying out the reduction step. A temperature of about 700° F. seems to combine a reasonably short reaction 81 time with the attainment of satisfactory magnetic properties. Where lower temperatures are used, for example 500° F., with longer reduction time, higher coercive force values result. Higher temperatures give lower coercive force values, and if the powdered material is held at a temperature of about 1200° F. in dry hydrogen for any length of time it will become overbaked. This overbaked material is characterized by a low coercive force, a high permeability and a high ratio of ferric induction to residual magnetism. The texture of such overbaked particles is rougher and the color grayer than that of good powder. During the reduction stage, the powder will be changed from a reddish or brownish color, depending on the oxide originally used, into a substantially black powder. At the point where the powder has almost turned completely black, the reduction is terminated and the powder cooled. Since the coercive force depends on the temperature, time, and atmosphere, these may be varied in order to control the final characteristics to come within desired limits.

Following the reduction stage, I have found it usually desirable to subject the material to a reoxidation step. This step consists in heating the reduced powder at temperatures in the range from about 400° to 900° F., and preferably from 500° to 700° F. in contact with the

atmosphere, or any oxygen containing atmosphere. In the course of this reaction, the black powder resulting from the reduction operation is
82 transformed into a tan or brown magnetic oxide having the empirical formula Fe_2O_3. Temperatures below the indicated range may be used, but the time necessary for any beneficiation to result from lower temperatures usually ranges into days or weeks. At temperatures in the range from 400° to 700° F., the conversion is completed in a few minutes, or not more than a few hours, depending on the starting material. While higher temperatures outside the prescribed range may also be used to decrease the required time, the resulting coercive force suffers from such higher temperatures. This value goes down with increased temperatures, slowly at first, and falls rapidly at temperatures between 900° and 1300° F. until the powder completely loses its magnetic qualities. This treatment at the higher temperatures may be used to control the magnetic properties to a desired specification. The reoxidation step is also dependent on the type of material employed since some finely divided synthetic powders are so reactive that they will change rapidly at low temperatures, and often with such violence that they burn.

The reoxidized powders have approximately the same magnetic properties as the black, reduced powder. However, the reoxidization step often results in increased uniformity, lower noise level when used for magnetic recording purposes, and better stability in the final product.

Examples of the preparation of various magnetic oxides in accordance with the method described in the present invention are given below:

83 EXAMPLE I

A finely divided Fe_3O_4 powder was prepared according to the manner similar to that outlined in U. S. Patent No. 2,133,267. This black powder had an average particle size of about 0.4 microns and had a cubic particle shape. As tested initially, the powder showed an H_c of 130 and a B_r of 500. The powder was placed in a container open to the atmosphere and heated at a temperature of 1300° F. for a few minutes. At the same time, the powder was agitated thoroughly so that every portion was exposed to the air while at this temperature. The product, after being cooled by dumping the same onto an iron plate, evidenced a red color rather than the original black. This red powder was placed in a closed vessel through which hydrogen was passed continuously while heating to a temperature of 600° F. for thirty minutes with thorough agitation. The vessel was then cooled in a cold water bath and the hydrogen cut off. The resulting powder was nearly black, but had a slight brownish tint. Physically, it had the same 0.4 micron cubic particles as the original material, but the H_c had increased to a value of 500 while the B_r was approximately 250. The material did not appear to be completely saturated at a field strength of 2000 oersteds, at which the measurements were made, so that the actual values were probably considerably higher.

In making the test of magnetic properties, the powder to be tested was mixed with a binder and coated onto a strip of cellulose acetate. The values given for coercive force are quite accurate, but the remanance measurements, being affected by the powder to binder ratio, the uniformity of the coating and like factors, must be taken only as approximations.

84

EXAMPLE II

In this experiment, the process of Example I was repeated except that the reduction step was carried out at a temperature of 650° F. for a period of 15 minutes. The resulting black product had an H_c of 440 and a B_r of 370. The magnetic powder was saturated with an impressed field of 2000 oersteds.

EXAMPLE III

In this experiment, the procedure of Example I was repeated except that the reduction was carried out at a temperature of 700° F. for a period of 10 minutes. The resulting black powder had an H_c of 360 and a B_r of 800 and was saturated at a field of

2000 oersteds.

EXAMPLE IV

The product of Example III given above was further treated by reoxidation at a temperature of 500° F. in an open vessel with thorough agitation to insure that all parts of the heated oxide were exposed to the air. The powder turned to a brown color, and when cooled it was found to have an H_c of 220 and a B_r of 520.

To illustrate the importance of an initially high temperature treatment, the following two experiments were run without the original treatment, or with the heat treatment being carried out at a lower temperature:

EXAMPLE V

The starting material used in Example I was placed in a hydrogen vessel and heated to a temperature of 700° F. with agitation as previously. When removed from the hydrogen chamber, the powder was still black and its magnetic properties showed an H_c of 150 and a B_r or 240.

EXAMPLE VI

The starting material used in Example I was placed in an open vessel and heated to a temperature of 400° F. for about one hour with agitation and air circulation. The color changed from black to brown, and magnetic tests showed an H_c of 150 and a B_r of 400.

EXAMPLE VII

A commercial practically non-magnetic hydrous ferric oxide powder having a particle size of about 0.030 microns was used as a starting material. The powder was heated to a temperature of 1300° F. with agitation in air. When cooled the color was dark brown, and magnetic tests showed it to have an H_c of 140 and a B_r of 200.

EXAMPLE VIII

The product of Example VII was heated and agitated in hydrogen at a temperature of 675° F. for 15 minutes. It was then cooled and removed with the usual precautions to prevent burning. The resulting powder was nearly black, with a reddish tint, and showed an H_c of 450, a B_r of 466, and an Ho value of 330.

86 EXAMPLE IX

The product of Example VIII was heated and agitated in air in an open container at a temperature of 700° F. When cooled, it had a dark red brown color and showed an H_c of 240 and a B_r of 242.

EXAMPLE X

The product of Example IX was treated again in hydrogen at 750° F. for five minutes. This treatment produced a black material with a reddish tint, having an H_c of 510, a B_r of 336 and an Ho of 400.

EXAMPLE XI

The product of Example IX was treated in hydrogen at 800° F. for five minutes. The resulting black powder had an H_c of 320 and a B_r of 480.

EXAMPLE XII

The product of Example VII was treated in hydrogen at 800° F. for five minutes, yielding a black powder having an H_c of 350 and a B_r of 480.

EXAMPLE XIII

The starting material used in Example VII was heated in air to 1650° F. and afterwards treated with hydrogen at 750° F. for thirty minutes. The resulting powder had an H_c of 275, a B_r of 500 and an Ho of 130.

87 EXAMPLE XIV

The starting material of Example VII was treated by heating and agitating in hydrogen at a temperature of 750° F. for thirty minutes. The product produced in this manner had an H_c of 150, a B_r of 150 and an Ho of 100, illustrating that the preliminary high temperature treatment is necessary to produce any distinct increase in coercive force.

EXAMPLE XV

Finely divided hydrous ferric oxides having a particle size of about 0.03 microns were subjected to a series of treatments as outlined in the previous examples.

The results obtained are tabulated below:

First Treatment	Second Treatment	Third Treatment	Fourth Treatment	Fifth Treatment	Color	Magnetic Properties	
						H_c	B_r
1500° F. air	800° F. hydrogen	-	-	-	Blk.	260	346
1500° F. air	800° F. hydrogen	650° F. air	-	-	Brn.	160	280
1500° F. air	800° F. hydrogen	650° F. air	800° F. hydrogen	-	Blk.	290	440
1500° F. air	800° F. hydrogen	650° F. air	800° F. hydrogen	650° F. air	Brn.	190	260

A variety of oxides can be treated in the manner shown in examples. These include oxides such as Fe_2O_3, Fe_3O_4, $Fe_2O_3.H_2O$ and other iron oxides which contain a trivalent iron atom. The process of the present invention is especially applicable to particles having sizes from a fraction of a micron in maximum dimensions to ten microns, and to particles
88 whose shapes are cubic, acicular, scales, irregular grains, etc. In practically all cases the magnetic properties of the oxides were improved substantially. In some cases, the procedure discussed in the present specification was the only one which had any effect whatsoever on increasing the magnetic properties of the particles.

From the foregoing it will be apparent that I have herein provided a method for increasing the magnetic properties of ferric oxides to make them amenable for use in magnetic impulse record members. The process has general applicability to all types of iron oxides, regardless of their particle size or shape of the crystals.

It will be understood that modifications and variations may be effected without departing from the scope of the novel concepts of the present invention.

89 I CLAIM AS MY INVENTION:

* * * * * *

PLAINTIFF'S EXHIBIT NO. 1
(Paper No. 15)

[Received, Patent Office, Nov. 19, 1954]

AMENDMENT "F"

153 SIR:

In response to the Office action dated July 2, 1954, applicant hereby amends as follows:

In the Claims:

Kindly cancel claims 17 and 30-34, inclusive.

Kindly add the following claims:

--35. In a method of making permanent magnet material from a starting synthetic iron oxide containing a trivalent iron atom having a coercive force less than 200 oersteds and having a particle size of one micron and less by reducing said iron oxide at an elevated temperature, the improvement which comprises the preliminary step of heating such starting synthetic oxide to a temperature between 1000° and 1750° F. for a sufficient length of time that when subsequently reduced at an elevated temperature below that used in said preliminary heating step said iron oxide acquires a coercive force value in excess of 200 oersteds and higher than could be obtained by employing the same reducing conditions in the absence of said preliminary heating step.--

154 --36. The method defined by claim 35 in which the preliminary heating step is carried on in the presence of an oxidizing atmosphere, and the subsequent reduction is carried out in a reducing atmosphere at a temperature of between 400 and 1200° F.--

--37. The method defined by claim 35 in which the starting synthetic oxide is of cubic particle shape, the preliminary heating step is carried out in the presence of air, the subsequent reduction step is carried out in a hydrogen atmosphere and the resulting reduced iron oxide has a coercive force valve in excess of 300 oersteds.--

--38. The method defined by claim 35 in which the starting synthetic oxide is ferrosoferic oxide (Fe_3O_4) of cubic particle shape, the preliminary heating step is carried out in the presence of air, the reduction step is carried out in a reducing atmosphere and the resulting reduced iron oxide is re-oxidized at a temperature from about 400° to 900° F. in an oxidizing atmosphere to produce an iron oxide having a coercive force value in excess of 200 oersteds.--

--39. Permanent magnet material adapted to form an element of a magnetic impulse record member consisting of synthetic magnetic iron oxide particles of a cubic particle shape less than one micron in size and having a coercive force value (H_c) between 200 and 450 and a remanence (B_r) of 200 and above.--

* * * * * *

161 STATE OF ILLINOIS)
COUNTY OF COOK) SS.

MARVIN CAMRAS, being duly sworn on oath, doth depose and say:

That he is the applicant named in the above identified application;

That he carried out the experiments set forth in Examples I through XV in said application and that said examples accurately state the conditions and observations that he maintained and made during such experiments;

That the experiment of Example I started with a black ferrosoferric oxide powder having magnetic properties, including an H_c of 130, which are not suitable for use in a modern magnetic impulse record member; that by heating the powder exposed to the atmosphere to a temperature of 1300° F. for a few minutes, a substantially non-magnetic, alpha iron oxide was obtained; and that upon reducing the latter oxide in a hydrogen atmosphere at a temperature of 600° F. for thirty minutes a product was obtained having an H_c of 500;

That, as shown in Example V, where the starting material of Example I was used and the preliminary heating to 1300° F. omitted, but the starting material was reduced in a hydrogen atmosphere at 700° F., the resulting powder had an H_c of only 150, compared with 500 for

the product of Example I; that this shows that the preliminary heating to
a temperature of between 1000 and 1750° F., as in Example I, was es-
162 sential to the production of a product having an H_c of at least 200
oersteds and therefore suitable for use in a magnetic impulse record
member;

That in the method of Example VII, a practically non-magnetic hydrous ferric oxide powder was heated to 1300° F. in the air to give a product, when cool, which was dark brown and had an H_c of 140, thereby indicating that the product had some permanent magnet properties but not such as would adapt it for use directly in a magnetic impulse record member; that when the material of Example VII was reduced in hydrogen at a temperature of 675° F. for fifteen minutes, as an Example VIII, the resulting powder had an H_c of 450, which value dropped to 240 upon reoxidation at 700° F., as in Example VIII, but rose to 510 upon re-reduction at 750° F. for five minutes, as in Example X, thereby showing that the starting material of Example VII, although different in its magnetic behavior upon being heated to a temperature of 1300 F., nevertheless was put in a condition by such pre-heating so as to be susceptible to satisfactory improvement in its permanent magnet properties upon reduction, or upon reduction followed by reoxidation;

That in the experiment of Example XIII, the starting material of
Example VII was preheated to 1650° F. and then reduced in a hydrogen
atmosphere at 750° F. for thirty minutes, giving a product having an
H_c of 275; whereas, as shown in Example XIV, if the same starting ma-
terial of Example VII was not subjected to the preheating step, but was
reduced in a hydrogen atmosphere at 750° F. for thirty minutes, the
resulting product had an H_c of only 150, as against the value of 275 for
the product from the same starting material but subjected to preheating
at 1650° F., thereby establishing the essentiality of the preheating
163 step to a temperature of between 1000 and 1750° F. in order to
obtain satisfactory magnetic properties in the reduced and reoxidized
product;

That from the experiments forming the basis for Examples I to

XV, inclusive, affiant concludes that in the case of iron oxides having a trivalent iron atom and a particle size of the order of one micron and less, a material having satisfactory magnetic properties cannot be obtained (unless the starting iron oxide is of acicular crystalline form) merely by reducing the starting material in hydrogen at an elevated temperature, or by first reducing and then reoxidizing; that in the case of the acicular Fe_2O_3 constituting the preferred starting material for the production of Fe_3O_4 or gamma Fe_2O_3 having a coercive force of at least 200 oersteds, as disclosed in affiant's earlier application Serial No. 763,486, satisfactory magnetic properties can be obtained without the preliminary heating step of the present method, but that in the case of iron oxide of other than acicular form and of a particle size of one micron and less it is necessary to preheat such iron oxide before the same is capable of being converted into high coercive force material by reduction or by reduction followed by oxidation; that even in the case of the preferred starting material of said application Serial No. 763,486, which is of elongated or acicular form, affiant has found some improvement, as evidenced by the results of the following experiment which was performed by affiant: 164

"An acicular yellow oxide (Fe_2O_3) was reduced at 700° F. in a hydrogen atmosphere to produce a black iron oxide (Fe_3O_4), which, when tested, had an H_c=330, and a B_r=235.

"Some of the same acicular yellow oxide was then heated in the presence of air to 1300° F. and reduced at 700° F. in a hydrogen atmosphere under identical conditions to produce a black iron oxide (Fe_3O_4) which, when tested, had an H_c=370, and a B_r=300."

That at the time of making the invention disclosed in said application Serial No. 763,486, affiant knew that if magnetic iron oxides were heated above a certain critical temperature, they would lose their magnetic properties; that affiant's recognition of this fact is apparent from the statement on page 11 of said application Serial No. 763,486, reading as follows:

"When the temperature is raised to about 1050° F. the magnetic properties fall off rapidly and are entirely lost at about 1200° F.";

That such knowledge, however, gave affiant no reason to suspect that a preliminary heating of an iron oxide of cubic particle form to a temperature of from 1000 to 1750° F. would enhance the magnetic properties obtainable by reduction, or by reduction followed by oxidation; that it was only when affiant made the experiments forming the basis of Ex-
165 amples I to XV of the instant application that he discovered in the case of iron oxides of other than acicular crystalline form and of less than about one micron maximum dimension, the preliminary heating step to between 1000 and 1750° F. was necessary in order to produce products from such starting material that had as good magnetic properties, or nearly as good properties, as those of the ferrosoferric and gamma ferric oxides of said application Serial No. 763,486;

That it is not, as the Examiner seems to suggest, a mere inclusion of a conventional step in the process claimed in application Serial No. 763,486, since the process of this application will not produce products of satisfactory magnetic properties without the inclusion of the preliminary heating step when the starting material is of other than acicular crystalline form and of a maximum particle size of around one micron or less; also, that while the magnetic properties of the final product of the instant application are "no better than those claimed in application Serial No. 763,486, the Examiner, in pointing this out, overlooks the fact that if it were not for the inclusion of the preliminary heating step to a temperature of from 1000° to 1750° F., the starting material of the instant application could not be converted into products having anywhere near as good magnetic properties as the products of the aforesaid application Serial No. 763,486;

That with respect to the product defined by claim 39 of the instant application, affiant believes it to be novel for a magnetic iron oxide of cubic particle shape and a particle size less than one micron to have an H_c value between 200 and 450; that he knows of no prior product of these

physical characteristics that possessed anywhere near the H_c value of
166 200 and that therefore the prior iron oxides of cubic particle form and of a size less than one micron were not satisfactory for use in present day, high fidelity magnetic impulse record members, whereas the product of claim 39 is suitable for such use;

That the present method enables the satisfactory use of a heretofore unsatisfactory source material for the making of modern sound tapes; that although ferrosoferric and gamma ferric oxides of satisfactory magnetic properties were produced by the method of affiant's prior applications, this may have been possible largely because of the elongated or acicular form of the particles and to the fact that such elongated particles are of a single magnetic domain type; and that therefore it was wholly unobvious that by a preliminary high heating step, old in itself, iron oxides of other than acicular crystalline form and less than one micron in maximum dimension could be converted into satisfactory material for use in modern magnetic recording tapes;

That whereas the methods of affiant's prior applications, when applied to iron oxide of cubic particle shape, do not give material of the desired magnetic properties, the method of the instant application does, thereby making a different source material available for the purpose;

That in the coating of iron magnetic strip material with the magnetic material of claim 39 in admixture with a resinous binder, the cubic particle shape and the fine particle size (of from less than one micron down to colloidal size), permit the use of different techniques with respect to the weight ratio of magnetic particles to binder, the manner of application of the particles to the non-magnetic strip material
167 and the manner of compacting the coating after application; and that, therefore, there is a certain degree of criticality with respect to both the size and cubic shape of such particles in connection with their use in magnetic impulse record members;

And further affiant sayeth not.

/s/ Marvin Camras

[Jurat dated November 4, 1954.]

172 PLAINTIFF'S EXHIBIT NO. 1
(Paper No. 20)

[Received, U. S. Patent Office, March 1, 1955]

BRIEF ON APPEAL
TO THE BOARD OF APPEALS

This is an appeal from the final rejection by the Primary Examiner, dated July 2, 1954, of the claims in the above identified application for patent. Subsequent to the date of such final rejection, applicant filed a new set of claims, numbered claims 35 to 39, inclusive, cancelling all the previous claims, and while the Examiner in his action of December 15, 1954 did not then enter this amendment, he indicated that the amendment would be entered for purposes of appeal. Accordingly, this appeal is taken with respect to claims 35 to 39, inclusive, which read as follows:

--35. In a method of making permanent magnet material from a starting synthetic iron oxide containing a trivalent iron atom having a coercive force less than 200 oersteds and having a particle size of one micron and less by reducing said iron oxide at an elevated temperature, the improvement which comprises the preliminary step of heating such starting synthetic oxide to a temperature between 1000° and 1750° F. for a sufficient length of time that when subsequently reduced at an elevated temperature below that used in said preliminary heating step said iron oxide acquires a coercive force value in excess
173 of 200 oersteds and higher than could be obtained by employing the same reducing conditions in the absence of said preliminary heating step.--

--36. The method defined by claim 35 in which the preliminary heating step is carried on in the presence of an oxidizing atmosphere, and the subsequent reduction is carried out in a reducing atmosphere at a temperature of between 400 and 1200° F.--

--37. The method defined by claim 35 in which the starting synthetic oxide is of cubic particle shape, the preliminary heating step is carried out in the presence of air, the subsequent reduction step is carried out in a hydrogen atmosphere and the resulting reduced iron oxide has a coercive force value in excess of 300 oersteds.--

--38. The method defined by claim 35 in which the starting synthetic oxide is ferrosoferric oxide (Fe_3O_4) of cubic particle shape, the preliminary heating step is carried out in the presence of air, the reduction step is carried out in a reducing atmosphere and the resulting reduced iron oxide is re-oxidized at a temperature from about 400° to 900° F. in an oxidizing atmosphere to produce an iron oxide having a coercive force value in excess of 200 oersteds.--

--39. Permanent magnet material adapted to form an element of a magnetic impulse record member consisting of synthetic magnetic iron oxide particles of a cubic particle shape less than one micron in size and having a coercive force value (H_c) between 200 and 450 and a remanence (B_r) of 200 and above.--

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THE INVENTION

The invention of this application relates to a method for producing magnetic iron oxides, either ferrosoferric oxide, Fe_3O_4, or gamma ferric oxide, Fe_2O_3, for use in magnetic impulse record members such as tapes, ribbons and the like of non-magnetic material coated or impregnated with a track of the magnetic iron oxide material. For magnetic recording purposes, the iron oxide material should have a coercive force, H_c, above 200, and preferably from 220 to 450, and the remanence, Br, should be at least 200. Also, the B_r versus H curve should have low values of B_r below the H_o and rise rapidly and fairly linearly above the H_o point. A sharp saturation value is desirable from the standpoint of ease of erasing, with saturation preferably occurring between the field of 700 to 2000 oersteds and the lower values being preferred.

The present application is a continuation-in-part of application Serial No. 763,486, filed July 25, 1947, now issued into patent No. 2,694,656, dated November 16, 1954. That patent particularly stresses the preparation of magnetic iron oxides of less than 6 microns in greatest dimension and having an acicular crystalline form. However, a different technique is necessary to impart adequate magnetic properties to

particles that are not of acicular crystalline form and that are as small
as 1 micron or smaller in maximum dimension. The different technique,
or method, forms the subject matter of this application. While primarily
directed to the treatment of iron oxides of cubic particle shape, that is,
particles that are substantially equi-dimensional, or equant, the appli-
175 cation also discloses that all forms of magnetic iron oxides, includ-
ing the acicular form, are improved to some extent by the treatment dis-
closed, and that the treatment is essential in the case of iron oxides of
cubic particle shape, or of substantially equant particle form.

The starting material for the method of the instant invention is an iron oxide containing a trivalent iron atom, particularly alpha ferric oxide, Fe_2O_3, its hydrate, $Fe_2O_3H_2O$, and ferrosoferric oxide, Fe_3O_4. Prior to treatment by applicant's method, these iron oxides may have magnetic properties, but, in general, the coercive force values are so low as not to adapt the starting material for use in magnetic recording tapes, and the like. Furthermore, if these iron oxides used as starting materials were merely to be subjected to reduction, or reduction and oxidation steps such as described and claimed in applicant's patent No. 2,694,656, their magnetic properties could not be sufficiently improved to render them suitable for magnetic recording purposes.

The present invention includes as the first step of the method the
heating of the starting iron oxides to an elevated temperature in the range
of from about 1000 to 1750° F., and normally between 1100° F. and 1500°
F. This preliminary heating step conditions the iron oxides in a way not
wholly understood so that upon subsequent reduction, or reduction fol-
lowed by oxidation, in the manner disclosed in this application and
described and claimed in patent No. 2,694,656, magnetic properties
are imparted to the iron oxides that could not otherwise be acquired
176 by them if the preliminary heating step were omitted.

The application contains fifteen examples, illustrating the use of the method in the treatment of various starting materials. In Example I, black ferrosoferric oxide, Fe_3O_4, having a cubic particle shape and an average particle size of about 0.4 micron was prepared in accordance with the method described in U. S. patent No. 2,133, 267. As so prepared, the powder exhibited a coercive force of 130 oersteds and a remanence of 500. This powder was then placed in an open container and heated at a temperature of 1300° F. for a few minutes, while being agitated thoroughly in exposure to the air. After being cooled, the resulting powder had a red color and was substantially non-magnetic. This red powder was then reduced at a temperature of 600° F. in a hydrogen atmosphere for thirty minutes with thorough agitation. After being cooled, the resulting powder was nearly black, but had a slight brownish tint. Physically it had the same 0.4 micron size cubic particles as the original material, but the coercive force value had increased to 500 oersteds, while the remanence was approximately 250.

Where the starting material is the same as in Example I, but, as in Example V, the preliminary heating to 1300° F. is omitted, the resulting powder after reduction in a hydrogen atmosphere at 700° F. had a coercive force, H_c, of only 150, compared with 500 for the product of Example I. This shows the essentiality of the preliminary heating step in the method of this application. This and other features of the
177 present invention are more specifically set forth and explained in the affidavit of applicant, accompanying the amendment filed November 19, 1954.

THE GROUNDS OF REJECTION

The claims were rejected as unpatentable over the claims of Serial No. 771,495, which application and its companion application Serial No. 763,486 were combined and issued as patent No. 2,694,656, in view of the following:

Welo and Baudisch article (1925), referred to as Baudisch.
Mellor "Treatise" (1934).
Abraham et al "Nature" (1925).

The Examiner has taken the position that the preliminary heating step called for by the claims on appeal is merely a conventional step of preparing alpha Fe_2O_3 or other iron oxide having substantially no magnetic properties.

In the Office action of December 15, 1954, the Examiner seems to base his final rejection largely upon his contention that the recitations in the claims with respect to the obtention of a higher coercive force in the ultimate iron oxide product when the preliminary heating step is utilized than when it is not utilized are not claimed positively and are "no more than an expression of a desired result." The Examiner further compares certain of the claims of this application with claims of the
178 issued patent and alleges that there is no patentable distinction between them.

THE CLAIMS SHOULD BE HELD PATENTABLE

With regard to the Examiner's contention that the claims do not positively set forth the distinctions urged by applicant, this is respectfully submitted to be an erroneous and untenable position. The rejection is particularly applied against claim 35, but since claims 36 to 38 are dependent thereon, it also applies to these claims. Claim 35 sets forth in the preamble the process to which the improvement applies, namely:

> "In a method of making permanent magnet material from a starting synthetic iron oxide containing a trivalent iron atom having a coercive force less than 200 oersteds and having a particle size of 1 micron and less by reducing said iron oxide at an elevated temperature * * *".

The claim then goes on to recite:

> " . . . . the improvement which comprises the preliminary step of heating such starting synthetic oxide to a temperature between 1000° and 1750° F. for a sufficient length of time that when subsequently reduced at an elevated temperature below that used in said preliminary heating step said iron oxide acquires a coercive force value in excess of 200 oersteds and higher than could be obtained by employing the same reducing conditions in the absence of said preliminary heating step."

In holding that the recitations in claim 35, line 9, beginning with "that when * * *" are no more than an expression of a desired result, the Examiner apparently assumes that the subsequent reducing step is

not positively claimed. However, the method to which the improvement
applies, as called for by the preamble, is one in which the starting synthetic
iron oxide is reduced at an elevated temperature. Thus, the reduction
step is positively claimed, and the comparison is made between
179 the coercive force values of the resulting product, both made from
the same starting synthetic iron oxide but the one starting synthetic
oxide being preliminarily heated to a temperature between 1000 and 1750°
F. for a sufficient length of time to acquire a coercive force value in
excess of 200 oersteds upon reduction, and the other not having been
subjected to such preliminary heating step.

The purpose of a claim is to "particularly point out and distinctly claim the part, improvement or combination which he (applicant) claims as his invention or discovery." Claim 35, it is respectfully submitted, conforms in every way to this requirement. In the preamble, the essential (but old in itself) step of reducing the iron oxide at an elevated temperature is recited, and then follows the improvement which comprises the preliminary step of heating the starting synthetic oxide to a temperature between 1000 and 1750° F. The period of time over which this preliminary heating step is carried out is defind as "a sufficient length of time (such) that when subsequently reduced at an elevated temperature below that used in said preliminary heating step said iron oxide acquires a coercive force value in excess of 200 oersteds and higher than could be obtained by employing the same reducing conditions in the absence of said preliminary heating step."

This is not a statement of a desired result, as the Examiner con-
tends, but is a criterion for the length of time of the preliminary heating
180 step. There has been no rejection of the claim on the ground of functionality,
nor should there be, since the test of coercive force value is
a simple one that can be easily made, so that the length of time can be
easily determined.

Furthermore, since the novelty does not reside in the reducing step, per se, nor in the preliminary heating step, per se, but in the combination of the two and in the particular sequence in which they are carried out, the claim should not be rejected on the ground that either of the steps, taken alone, is old. The sole novelty here resides in the sequence of steps, rather than the individual steps. As was held by the Board of Appeals in Ex parte Schwenk, Bloch and Whitman, 73 USPQ 85, 86:

> "If the sequence of steps is broadly new, and this appears to be the case, and the individual steps themselves are old in this particular branch of chemistry and properly identified in the specification, it is our view that the manner in which the individual steps is carried out need not be specifically included in the claim where, as here, the sole novelty appears to reside in the sequence rather than in the individual steps themselves."

None of the references discloses the sequence of steps here recited, first a preliminary heating step to condition the starting synthetic oxide so that it will be capable of acquiring a higher coercive force value than could be obtained without the preliminary heating step, and secondly, a reduction step at an elevated temperature.

Possibly, the Examiner's rejection, although not so stated, is on the ground that the reducing step is not positively included. The reducing step, however, is included in the preamble, and under the authority of the decision of the Board of Appeals in Ex parte Purington, 64 USPQ 202, the preamble should be considered as a material part of the claim. 181 It recites the method in which the preliminary heating step is included as an improvement.

As already pointed out, certain of the examples of the specification establish the improvement in coercive force values that can be made by the use of the preliminary heating step, and the fact that higher coercive force values can be obtained by such use of the preliminary heating step than can be obtained without it. The Honorable Board's careful attention is directed to the affidavit of applicant accompanying the amendment filed November 19, 1954. Reference has already been made to Examples I and V and to the fact that as a result of the use of the preliminary heating step, a coercive force value of 500 was obtained, as against only 150 without the preliminary heating step. Similarly, with respect to Examples VII, VIII and X, a practically non-magnetic hydrous ferric oxide powder was heated in accordance with Example VII to a temperature of 1300° F. to give a product having an H_c of 140, --too low for use directly in a magnetic impulse record member--, but after reduction in accordance with Example VIII the resulting powder had an H_c of 450, which value dropped to 240 upon reoxidation at 700° F. as in Example VIII but rose to 510 upon re-reduction at 750° F. for five minutes, as in Example X. This series of examples shows that the starting material of Example VII was put in condition by the preheating step so as to be susceptible to satisfactory improvement in its permanent magnet properties upon reduction, or upon reduction followed by re-oxidation.

Again, when the starting material of Example VII was preheated to 1650° F., as in Example XIII, and then reduced in a hydrogen atmosphere
182 at 750° F. for thirty minutes, it gave a product having an H_c of 275; whereas, as shown in Example XIV, if the same starting material of Example VII was not subjected to the preheating step, but was directly reduced in a hydrogen atmosphere at 750° F. for thirty minutes, the resulting product had an H_c of only 150. This establishes the essentiality of the preheating step to a temperature of between 1000 and 1750° F. in order to obtain satisfactory magnetic properties in the reduced or the reduced and reoxidized product.

It also establishes the validity of the comparison of products as to coercive force values, when using the preliminary heating step as against not using the preliminary heating step. Consequently, claim 35 is properly based upon the specification and sets forth a criterion as to what constitutes the "improvement" which can be easily measured by comparison of coercive force values obtained, first by employing the method set forth in the claim that includes the preliminary heating step, and, secondly, by employing a method not covered by the claim in which the preliminary heating step is omitted.

The Examiner refers to claim 21 of patent No. 2,694,656 as emphasizing what he terms "the evanescent differences between applicant's prior application Serial No. 771,495 coupled with the art as applied in the final rejection and the present case." Claim 21 of the patent is quoted for convenience, as follows:

> "21. The method of making permanent magnet material,
> which comprises precipitating crystals of $Fe_2O_3H_2O$ from solution
> of a particle size of less than 6 microns in greatest dimension,
> heating said crystals in the air to a temperature between 500 and
> 1550° F. to drive off the water of hydration, reducing said de-
> hydrated crystals at a temperature between 500 and 1000° F. in
> a gaseous reducing atmosphere and cooling in a gaseous reducing
> 183 atmosphere to obtain a magnetic material also in crystalline form
> and of a particle size less than 6 microns and consisting essen-
> tially of Fe_3O_4 particles having a coercive force value of at least
> 200 oersteds and a B_{fm}/B_r (H=1000) of not over 3 to 1."

The above quoted claim is specific to the purpose for which the ferric oxide monohydrate is heated, namely "to drive off the water of hydration". In the absence of the teachings of the present application, there would be no reason for the preliminary heating step to drive off water of crystallization unless the starting material was hydrated, and even if it were hydrated, there would be no assurance that the temperatures and times employed for driving off the water of hydration would also necessarily and inherently enhance the susceptibility of the iron oxide to acquire a higher coercive force value than could be obtained without the preliminary heating step. Present claim 35 could not properly have been made in applicant's earlier application Serial No. 771,495. In rejecting the claim of this application on claim 21 of the patent, the Examiner falls back upon the disclosure in the prior art of the old step of heating magnetic iron oxide above the Curie point, or some critical temperature, to render it non-magnetic, but such prior art does not teach or even suggest the combination of a preliminary heating step with a subsequent reducing step to improve the permanent magnet properties of the resulting product. It is in the sequence of steps that patentability of claims 35 to 38, inclusive, resides, and not in the single step of preheating or in the subsequent step, by itself, of reducing at an elevated temperature.

That the sequence of steps is the essential thing is entirely evi-
184 dent, since if the order of steps were reversed, the final product, following the heating step at 1000 to 1750° F., would have no or very inferior magnetic properties. The present claims cover only a method in which the preliminary heating step is employed and in which, as a result of the preliminary heating step, higher coercive force values are obtained after reduction, or after reduction and oxidation, than could have been obtained if the preliminary heating step were omitted. In the then copending application Serial No. 771,495, the starting material is of acicular crystalline form and the preliminary heating step is not necessary to attain coercive force values in excess of 200 oersteds. That is apparent from the fact that no preliminary heating step is even disclosed, except for the purpose of eliminating water of hydration. While there may be, as set forth in the affidavit of applicant, some improvement in coercive force values even in the case of an acicular crystalline form of iron oxide starting material, the present method claims do not cover the use of the preliminary heating step in all instances, but only where higher coercive force values are obtained than could be obtained without such preliminary heating step.

With regard to the Examiner's contention that the magnetic properties here claimed are no better than those claimed in Serial No. 771,495, this is submitted to be immaterial. The point is that the present method enables the obtaining of coercive force values in excess of 200, starting with iron oxides of a particle form from which materials having such coercive force values could not otherwise be obtained. Thus,
185 the present method makes available to the magnetic impulse record industry another source of permanent magnet material suited for such impulse record members, not heretofore available for that purpose. This, in itself, is a contribution to the art for which applicant should be awarded patent protection.

Granted that the prior art recognized the heating step, per se, as a conventional step of preparing alpha Fe_2O_3 or an iron oxide having substantially no magnetic properties, where is the teaching in the prior art that by first so destroying the magnetic properties, one can render the iron oxide more susceptible to the acquisition of higher permanent magnet properties in the subsequent reducing step, or reducing followed by an oxidizing step? That, it is respectfully submitted, is the test, rather than is the preliminary heating step, taken by itself, old?

With regard to claim 39, the Examiner compares that claim with claim 25 of patent No. 2,694,656. Claim 25 of the patent reads as follows:

> "25. Ferromagnetic iron oxide selected from the group consisting of a synthetic ferrosoferric oxide, Fe_3O_4, and of a synthetic gamma ferric oxide, Fe_2O_3, adapted to form an element of a magnetic impulse record member, said iron oxide consisting essentially of uniformly small elongated crystals of less than about 1.5 microns maximum dimension having a length-to-width ratio of about 2.5 to 1 and higher, and having a cubic crystal lattice structure and a coercive force, H_c, within the range of 245 to 330 and remanence, B_r, of above about 500 gauss."

The above quoted claim 25 of the patent is directed to elongated crystals, whereas claim 39 is restricted to synthetic magnetic iron oxide particles of a cubic particle shape. This reference to cubic particle 186 shape should not be confused with the reference in claim 25 to "a cubic crystal lattice structure", since the former is an exterior characteristic, whereas the latter is a characteristic of the internal atomic structure.

As previously pointed out, it is immaterial that the product of the patent has as high or higher coercive force values and higher remanence values. The significant thing is that in the case of cubic particles it was impossible prior to the present invention to impart to such cubic particle shaped iron oxides a coercive force value as high as 200. Applicant has repeated a great many procedures given in the prior art for the making of permanent magnet iron oxide allegedly suitable for use in magnetic impulse record members and has invariably been unable to produce coercive force values in excess of about 140 oersteds. The Examiner, instead of comparing the coercive force values of applicant's cubic shaped particles with coercive force values of prior art material also of cubic particle shape, compares these values with a value set forth in applicant's prior patent where the particles are of acicular or elongated shape and where coercive force values in excess of 200 can be realized without the preliminary heating step.

The Examiner also fails to give any significance to the particle size recited in claim 39, namely of less than 1 micron in size. As established by the affidavit previously referred to, the cubic particle shape and the fine particle size, namely, of from less than 1 micron down to colloidal size, permit the use of different techniques with respect to the weight ratio of magnetic particles to binder, the manner of application
187 of the particles to the non-magnetic strip material and the manner of compacting the coating after application. There is, therefore, a certain degree of criticality with respect to both the size and cubic shape of such particles in connection with their use in magnetic impulse record members.

With the emphasis that is now being placed upon high fidelity magnetic impulse record members, it seems unrealistic to say the least not to give patentable significance to the permanent magnet properties of the product defined by claim 39 that peculiarly adapt such product for use in magnetic impulse record members, where similar iron oxides of cubic particle shape were not so adapted. Furthermore, the Examiner is now being inconsistent in refusing the allowance of claim 39 on the ground of unpatentability over claim 25 of the aforesaid patent, since, obviously, claim 25 was allowed on the basis of the combination of a particular particle shape, acicular, and a particular set of permanent magnet characteristics. Just as the permanent magnet characteristics cited in patented claim 25 distinguish the acicular form of iron oxides from theretofore known acicular iron oxides, so, here, the magnetic characteristics recited in claim 39 distinguish the cubic particle form of iron oxides claimed from cubic-shaped iron oxide particles known to the prior art.

188 CONCLUSIONS

In view of the foregoing explanation as to the differences between the claims on appeal and the methods and products disclosed in the prior art and claimed in applicant's patent No. 2,694,656, it is respectfully submitted that all of the claims define novel and patentable subject matter and should be allowed. The allowance of the claims and the reversal of the Examiner's holding with respect thereto are earnestly solicited.

An oral hearing is respectfully requested.

Very respectfully,
THE FIRM OF
/s/ Charles W. Hills
Attorneys for Applicant

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190 PLAINTIFF'S EXHIBIT NO. 1
(Paper No. 21)

[Mailed May 25, 1956]

EXAMINER'S ANSWER

This is an appeal from the final rejection of claims 17 and 30-34. The amendment and affidavit filed November 19, 1954 (Paper No. 15) which presents new claims 35-39 and cancels claims 17 and 30-34 has been entered for purposes of appeal in accordance with the examiner's letter of December 15, 1954 (Paper No. 16). The only claims in the case are, therefore, claims 35-39.

The Claims

The claims are correctly reproduced in applicant's brief, pages 1 and 2.

The Invention

Applicant's discussion of the contended invention on pages 3-6 of his brief appears to be adequate.

It is to be noted that applicant's patent 2,694,656 indicates that
the invention therein described is directed to treating an iron oxide
preferably acicular in crystal form; however, other forms can be pro-
duced having in general, similarly desirable magnetic properties, column
2, lines 13-16 and that the iron oxide starting material used may be in
191 the form of uniformly small crystals having a length of the order of
1 to 2 microns, column 2, lines 19-23 or smaller than 1 micron, column
3, lines 38-43. The specification indicates the present contended in-
vention is applicable to iron oxide starting materials, regardless of the
form of the individual particles, page 3, lines 6-9, 19-26, page 13,
last five lines, page 14, lines 1-4 including cubic particles, page 3,
lines 10-18 and page 14, line 1.

It is to be noted that Examples I and V in applicant's specification are not conducted under the same process conditions. The reduction step of Example V is conducted at a higher temperature than the temperature used in Example I and as indicated in applicant's patent 2,694,656 the coercive force (Hc) of the ultimate magnetic iron oxide decreases with increase in reduction temperature, column 4, Table I. Example V does not state the time during which reduction was in progress and as indicated in applicant's patent 2,694,656, column 5, Table II, the coercive force (Hc) of the ultimate magnetic iron oxide obtained decreases with increase in reduction time.

The References

The references are listed in applicant's brief, page 6. Discussion thereof will be limited in the interests of brevity to the Examiner's discussion of the final rejection of claims 17 and 30-34.

The Rejection

Claims 30-34 were rejected as unpatentable over the claims of applicant's copending application S. N. 771,495, now patent 2,694,656, hereinafter referred to as Camras, in view of Baudisch, Mellor or Abrahams et al. New claims 35-38 were subsequently (Paper No. 16) similarly so considered.

192 Claims 35-38 require that a starting synthetic iron oxide (not necessarily one of cubic particle shape, claims 35 and 36) be "conditioned" or preliminarily heated in air to a temperature between 1000° and 1750° F. Thereafter the "conditioned" iron oxide is treated in the same manner as any iron oxide starting material would be in accordance with the claims of Camras, e.g. claim 13 or 19 with respect to a subsequent reduction step and claims 20 or 24 with respect to subsequent reduction and oxidation steps to form a magnetic iron oxide having a coercive force value in excess of 200 oersteds.

The contended distinction of conducting a preliminary heating or oxidation step at 1000-1750° F. with respect to an iron oxide having a coercive force less than 200 oersteds was considered a conventional way of obtaining a non-magnetic iron oxide which the claims of Camras require, e.g. claim 13, "providing a synthetic non-magnetic oxide . . ." and was shown to be conventional by any one of Baudisch, Mellor or Abraham et al who teach that alpha ferric oxide (non-magnetic) may be prepared from a magnetic iron oxide having a trivalent iron atom by heating the magnetic iron oxide in air at temperatures of the order claimed. No difference in ultimate result was apparent, that is, a magnetic iron oxide having a coercive force of above 200 oersteds is obtained. No critical difference was considered made out by the specification with respect to "conditioning" a cubic shaped particle of iron oxide over any other shaped particle, either in process requirements or ultimate result. Claims 35 and 36 further indicate that the process claimed is not critical to "cubic" shaped iron oxide. In any event, utilizing a known method of obtaining a non-magnetic iron oxide such as taught by Baudisch, Mellor or Abraham et al as required by the claims of Camras was considered not to involve invention.

193 The new claims 35-38 were further indicated to not distinguish over claim 21 of Camras. Claim 21 of Camras provides for a preliminary "heating" step in which ferric oxide monohydrate is heated in air at a temperature between 500-1550° F. Procedurally any distinction between the step of Camras in combination with a subsequent reduction

step and the preliminary "heating" step of claims 35-38 in combination with subsequent reduction (claims 35-37) and oxidation step (claim 38) was considered nebulous.

Distinctions in terms of result desired were not considered limitative, and in any event not considered to differ in the results obtained by Camras. For example, in claim 35, the starting material is an iron oxide which contains a trivalent iron atom having a coercive force less than 200 oersteds. Obviously alpha ferric oxide (non-magnetic) such as the alpha ferric oxide of Camras is included in the claimed expression. Further, magnetic gamma ferric oxide or ferrosoferric oxide, such as those of Baudisch, Mellor or Abraham et al prior to the heating of such oxides to render them non-magnetic, is included in the claimed expression. The magnetic iron oxide obtained by Camras by a reducing step with or without an oxidation step at temperatures below those indicated by Baudisch, Mellor or Abraham et al necessary to obtain a non-magnetic ferric oxide and at a temperature below the maximum (1500° F.) stated in claim 21 of Camras acquires a coercive force in excess of 200 oersteds and higher e.g., 550 oersteds, column 1, line 45 of Camras.

The affidavit of applicant (Paper No. 15) was carefully considered. It sets forth essentially the same data that appears in the specification in Examples I, V, VII, VIII. The remainder of the affidavit merely states applicant's conclusions. The affidavit is self-serving.

194 Examples I and V have been discussed supra.

Examples VII, VIII and X merely indicate, as does the patent to Camras and claims of Camras, that reduction of an iron oxide in the manner claimed may produce ferrosoferric oxide having a high coercive force (550 oersteds) and low temperature oxidation or reoxidation may produce gamma ferric oxide having a high coercive force (330 oersteds). As indicated by Baudisch, Mellor and Abraham et al the higher the temperature at which a magnetic oxide is heated the less magnetic the ultimate oxide and as indicated by Camras the higher the temperature of reduction, the less magnetic the oxide; the longer the reduction time, the less magnetic the oxide; the higher the oxidation (reoxidation) temperature the less magnetic the oxide. The examples, therefore, were not considered to clearly point out any distinguishing characteristics in result over Camras as modified by Baudisch, Mellor or Abraham et al.

Claim 17 was rejected as unpatentable over the product claims of Camras. Claim 39 was similarly so considered, noting claim 25 of Camras. The product claim 25 of Camras has (1) a coercive force of comparable value to the coercive force of claim 39 (2) a comparable particle size and (3) a higher remanence value. The only apparent distinction between claims 39 and the product claims of Camras is the particle shape. Although claim 25 may require an acicular particle shape, some other product claims, e.g. claims 4 and 5, do not positively require the magnetic iron-oxide material to be of acicular particle shape and in that sense are generic to all particle shapes of magnetic iron oxide. In any event, no criticality was considered to have been made out for the particular shape of particles claimed; the difference was considered one of degree.

195 For the reasons indicated above the final rejection is considered proper and affirmance of the examiner's position is respectfully requested.

Respectfully submitted,

/s/ B. H. Enkin
Examiner

/s/ CDQ
CDQuarforth/mpm

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196 PLAINTIFF'S EXHIBIT NO. 1
(Paper No. 22)

[Received, June 13, 1955, U. S. Patent Office]

REPLY BRIEF

The Examiner's Answer, dated May 25, 1955 in the above identified cause has been received and noted.

The purpose of this reply is to correct some erroneous conclusions of fact and conclusions of law made by the Examiner.

The Examiner cites the applicant's patent No. 2,694,656 as indicating that the invention there described may be used for treating an iron oxide of any form. He specifically refers to column 2, lines 13 to 16, which state:

> "In either case, the materials of my invention are preferably in acicular crystalline form, although other forms can be produced having, in general, similarly desirable magnetic properties."

Applicant is unable to understand how the specification of his copending application, which has now issued as patent No. 2,694,656 can adversely affect the patentability of the claims in the instant appeal. Clearly, the specification of that patent is not a reference and the Examiner's use of such specification is not justified or pertinent.

We shall not labor at length on the circuitous type of reasoning in which the Examiner has engaged in order to build up an anticipation of the applicant's appealed claims. The Examiner's difficulty apparently stems from the fact that he is unfamiliar with the practical art of making magnetic impulse record members. 197 The Examiner, for example, puts considerable emphasis upon the applicant's patented claims which, inter alia recite the step of "providing a synthetic nonmagnetic ferric oxide". Perhaps the Examiner does not realize that practically all synthetic ferric oxides as initially produced are nonmagnetic or at least too feebly magnetic to make them useful as powders for the production of magnetic impulse record members. To the Examiner, this step of "providing" the oxide inherently anticipates the applicant's unique discovery that the high temperature treatment prior to reducing the oxide conditions the oxide so that in the subsequent reduction reaction, a higher coercive force is obtained than could be obtained in the absence of the conditioning treatment.

We are frankly unable to see any logic in the Examiner's position that since the claims of applicant's patent require "providing a synthetic non-magnetic oxide" and since heating at 1000-1750° F. is "a conventional way of obtaining a non-magnetic iron oxide", the present claims do not involve invention. This reasoning might have some force if it were necessary in order to provide a synthetic non-magnetic oxide to go through a preliminary heating step such as herein claimed. The contrary, however, is the case. As we have already said most, if not all, synthetically produced iron oxides are initially non-magnetic, or only feebly magnetic. The precipitation method of Example I of the Camras patent is a typical method of producing alpha $Fe_2O_3 . H_2O$ -- a non-magnetic iron oxide. Why should one familiar with the patent and attempting to follow its teachings resort to some additional step, -- not disclosed therein, to "provide a non-magnetic oxide"? 198

Even more significantly, if one already has a synthetic iron oxide that is non-magnetic or only feebly so and wanted to increase its permanent magnetic properties, why would one deliberately heat such synthetic iron oxide to a temperature known to destroy magnetism in a magnetic iron oxide? Common sense, if nothing else would, in fact, contraindicate the applicant's unique conditioning step because, as we have repeatedly stated in the prosecution of this case, it is well known that many oxides of iron lose whatever magnetism they may have by treatment at elevated temperatures. Would it not be more logical to adopt a treating technique which would retain what small amount of magnetic properties the oxide already has rather than take a chance on destroying the inherent magnetic properties?

With regard to claim 21 of the Camras patent, the preliminary heating step there is specifically applied to $Fe_2O_3 . H_2O$ for the purpose of driving off water of hydration. Although the patent claim mentions

a temperature between 500 and 1550° F. for this purpose, the elimination of water of hydration could obviously be carried out at 500° F. or at a temperature below 1000° F., which is the minimum temperature set forth in the present claims. If, as recited in claim 21, the $Fe_2O_3 \cdot H_2O$ is obtained by precipitation, it is necessarily acicular and requires no preliminary heating to between 1000 and 1500 or 1750° F. to increase the magnetic properties obtained upon subsequent reduction in accordance with the further step recited in the claims. The present claims apply only to a synthetic non-magnetic or feebly magnetic iron oxide incapable, without the preliminary heating step, of acquiring permanent magnetic
199 properties represented by a coercivity in excess of 200 oersteds. There is, therefore, no overlapping between the present claims and patented claim 21.

Reconsideration of the Examiner's statement in the light of the foregoing comments is respectfully requested.

The allowance of the claims and the reversal of the Examiner's rejection with respect thereto are again earnestly solicited.

Very respectfully,
THE FIRM OF
/s/ Charles W. Hills
Attorneys for Applicant

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201 PLAINTIFF'S EXHIBIT NO. 1
(Paper No. 24)

[Mailed March 8, 1956]

DECISION OF THE BOARD OF APPEALS

Ex parte Marvin Camras

Application for Patent filed September 8, 1949, Serial No. 114,677.
Coating Material for Magnetic Recording.

Charles W. Hills for appellant.

This is an appeal from the action of the examiner in refusing to allow claims 35 to 39, inclusive, which replace the claims under final rejection.

Claims 35 and 39 are reproduced as illustrative.

35. In a method of making permanent magnet material from a starting synthetic iron oxide containing a trivalent iron atom having a coercive force less than 200 oersteds and having a particle size of one micron and less by reducing said iron oxide at an elevated temperature, the improvement which comprises the preliminary step of heating such starting synthetic oxide to a temperature between 1000° and 1750° F. for a sufficient length of time that when subsequently reduced at an elevated temperature below that used in said preliminary heating step said iron oxide acquires a coercive force value in excess of 200 oersteds and higher than could be obtained by employing the same reducing conditions in the absence of said preliminary heating step.

202 39. Permanent magnet material adapted to form an element of a magnetic impulse record member consisting of synthetic magnetic iron oxide particles of a cubic particle shape less than one micron in size and having a coercive force value (H_c) between 200 and 450 and a remanence (B_r) of 200 and above.

The references relied upon are:

Camras, 2,694,656, Nov. 16, 1954,

Abraham et al, "Nature", page 930, Vol. 115, No. 2902, June 13, 1925, Welo and Baudisch, "Two Stage Transformation of Magnetic into Hemotite" Phil. Mag., Vol. 50, pages 399-408, (1925),

Mellor, "Treatise on Inorganic and Theoretical Chemistry", Vol. 13, pages 732, 736, 738, 380, Pub. Longmans, Green & Co., New York City, 1934.

The appealed claims relate to an improvement in a method of preparing a magnetic iron oxide and to the product thus obtained. The method is said to be an improvement or modification of the method disclosed and claimed in appellant's prior patent, which issued from an application which was copending with the present application. The improvement comprises heating the iron oxide which constitutes the starting material to a temperature between 1000 and 1750° F. prior to the reducing step in a hydrogen atmosphere. The reducing step is conceded to be disclosed and claimed in appellant's prior patent.

Appealed claims 35 to 38 have been rejected as unpatentable over patented claim 13 in view of Mellor, Abraham et al or Welo and Baudisch. The examiner refers to the last mentioned citation as Baudisch but the publication is by the joint authors Welo and Baudisch and we will hereafter refer to it as Welo et al. Claims 35 to 38 were also rejected as unpatentable over claim 21 of the patent.

The auxillary references describe the transformation of magnetic
203 iron oxides to a nonmagnetic form of ferric oxide. Since all three of the references disclose substantially the same subject matter, and since Welo et al appears to be the most detailed, we will confine our attention to that citation.

Welo et al describe a number of experiments with magnetic iron oxide (Fe_3O_4). It is stated that this magnetic oxide may be oxidized to ferric oxide (Fe_2O_3) without changing its crystalline structure and without destroying its magnetic properties if the oxidation is carried out at low temperatures, the limiting temperature being approximately 550° C. Oxidation at higher temperatures or heating of the previously oxidized material at temperatures above 550° C. destroys the magnetic properties. It will be noted that the minimum heating temperature of

1000° F. specified in the appealed claims is just above the 550° C. mentioned by Welo et al as causing the transformation of the magnetic ferric oxide to the non-magnetic form. Patented claim 13 refers to "providing a synthetic non-magnetic ferric oxide in crystalline form and of a particle size not over 6 microns . . . " The examiner holds that no invention would be involved in employing conventional prior art practices to "provide" the nonmagnetic ferric oxide. Appellant insists that the exceptional improvement in magnetic qualities of the product by reasons of the claimed process warrants a holding that such claimed process is patentable over prior art. It is appellant's view that the preliminary step of patented claim 13 of "providing" a nonmagnetic ferric oxide is not suggestive of processes of the type described by Welo et al.

204 In rejecting claims 35 to 38 as without patentable merit over claim 21 of the patent, the examiner holds that the step of heating "in the air at a temperature between 500-1550°F." is the same as the preliminary step of heating recited in the claims under consideration. Appellant directs attention to the statement of purpose "to drive off the water of hydration" of patented claim 21 and insists that the heating step of the claims under consideration must be to a greater degree or for a greater time than is required for mere dehydration. We note that the specification of this case states that no particular heating time is essential, it being said that the desired change occurs "almost instantaneously as soon as the proper temperature is reached." It appears from Welo et al that the change to a nonmagnetic material occurs as a pure temperature effect as soon as the requisite temperature of at least 550° C. is reached. We are unable to appreciate how the heating step of claim 21 could be conducted within the portion of the claimed range running from 1000°F. to 1550°F. without also attaining the change here sought.

We have carefully studied all of the contentions presented on behalf of appellant but without finding reversible error in either of the foregoing rejections.

Appellant suggests that the patented process is directed primarily to the preparation of a magnetic material from an iron oxide in acicular crystalline form. It is further asserted that the presently claimed process is particularly adapted to the treatment of iron oxide of smaller 205 particle size and non-acicular form. The claims of the patent are not restricted to the treatment of an iron oxide in acicular form. The text of the patent refers to the process as applicable not only to the acicular form but also to other forms of iron oxide. Appellant protests the examiner's reference to the description of the patent since the rejection under consideration is as for double patenting but we do not believe that objection is sound. The rejection is as for double patenting. The reference to the specification is not for added subject matter not found in the claims but merely in further elucidation of the scope of the patented claims. In re Greenlee, 106 USPQ 104; 697 OG 556; 222 F.(2d) 739.

Appellant also referred to the present process as being especially applicable to small size particles. The patented claims refer to less than 6 microns. The presently considered claims refer to less than 1 micron. The column 3, lines 39 to 42 of the patent describes a product having crystals running from 1/4 to 1-1/2 microns. We find no patentable distinction based on the size of the particles.

Claims 36, 37 and 38 are dependent on claim 35. Appellant makes no serious contention that these claims are patentable in the absence of the allowability of claim 35.

Claim 39 defines the product. This claim has been rejected as without patentable merit over claim 25 of the patent. Appellant directs attention to the requirement for particles of cubic shape. The patented claim refers to elongated crystals having a length-to-width ratio of about 2.5 to 1 thus excluding cubic particles. The examiner held that 206 the record of this case did not justify a conclusion that the restriction to cubic particles was of inventive significance and directed attention to patented claim 4, which is silent as to the shape of the particles.

Appellant has discussed at some length the results set forth in the various examples of the specification and asserted that such examples show the unexpected superiority attained by the preheating step. In example 1 a magnetic oxide (Fe_3O_4) is employed and this material is first transformed into nonmagnetic ferric oxide by heating in air at a temperature of 1300°F. The material is then reduced in hydrogen in a manner corresponding to that disclosed and claimed in appellant's prior patent. The transformation to the nonmagnetic oxide is by the procedure disclosed in Welo et al. The further examples dealing with the same starting material show only that it is desirable to provide a nonmagnetic ferric oxide in following such processes as are defined in patented claim 13.

The decision of the examiner is affirmed.

/s/ E. W. Geniesse Examiner-in-Chief /s/ C. S. Duncombe Examiner-in-Chief /s/ I. Lisann Examiner-in-Chief (Acting)	BOARD OF APPEALS

March 8, 1956

Charles W. Hills
1414 Monadnock Block
Chicago 4, Illinois

207 PLAINTIFF'S EXHIBIT NO. 1
(Paper No. 25)

[Received, March 27, 1956, U. S. Patent Office]

PETITION FOR REHEARING OR RECONSIDERATION

In its decision dated March 8, 1956, the Board of Appeals affirmed the Examiner's rejection of all claims in the application. Reconsideration of the decision is requested or, in the alternative, a rehearing of the appeal is solicited.

The Board of Appeals affirmed the rejection of the method claims on the grounds of:

(1) Unpatentability over claim 13 of the copending Camras Patent No. 2,694,656 in view of the Welo et al article.

(2) Unpatentability over claim 21 of the aforementioned patent.

In rejecting the applicant's claims on the basis of claim 13 of the issued patent, the Board placed considerable emphasis upon the single word "providing" in the preamble of the patented claim which reads: "The method of making magnetic iron oxide which comprises providing a synthetic non-magnetic ferric oxide in crystalline form and of a particle size not over 6 microns in its greatest dimension and reducing said non-magnetic oxide at elevated temperature . . .".

We respectfully request the Board of Appeals to reconsider the language of claim 13 in the context of the specification from which it
208 was derived. The word "providing" as used in claim 13 of the patent is nothing more than a broad way of covering the various ways of obtaining the starting material for the subsequent reduction treatment. The interpretation of the word given by the Board of Appeals assumes the phrase "providing a synthetic non-magnetic ferric oxide" to have an exactly synonomous meaning with the phrase "demagnetizing a ferric oxide".

We submit that this interpretation does not find any basis in the descriptive matter of the specification of the patent. The non-magnetic properties of the crystals prepared according to the methods described in the specification of the patent are inherent by virtue of their mode of preparation; it is unavoidable that the starting ferric oxides are non-magnetic. There is no teaching in the specification or in the claims of the issued patent which would suggest to one skilled in the art the apparently contradictory thought of starting with a material which is already magnetic and then intentionally demagnetizing the material by high temperature treatment before proceeding to the reduction treatment to increase the coercive force and other desirable magnetic properties of the material. The far more logical approach, when starting with a material which has some magnetic properties, would be to immediately subject the material to a reduction treatment which, as pointed out by the issued patent, will greatly increase the coercive force of synthetic ferric oxides. Yet unless the ferric oxide starting material of the appealed claims is subjected to the heat treatment step before it is reduced, it will not acquire upon reduction a sufficiently high coercive force value (200 oersteds or over) to make it satisfactory for use in magnetic record members.

209 Consequently, the attempted combination with the claimed subject matter of the applicant's prior patent of the Welo et al article which merely states that a magnetic ferric oxide can be rendered non-magnetic by heating to high temperatures is neither obvious nor suggested by the article. To deliberately render non-magnetic an already magnetic iron oxide and then cause said oxide to regain magnetic properties by a subsequent step, would be contrary to scientific methods and contrary to common sense unless some advantage were known for the otherwise unnecessary step. Applicant has discovered that this type of pre-treatment will enable certain iron oxides which do not acquire sufficiently high permanent magnet properties in response to reduction treatments alone, to achieve coercive force values higher than could be obtained without the pretreatment and sufficiently high to make them usable as magnetic record members. Applicant has thus proceeded in a manner contrary to what the person having ordinary skill in the art would be expected to do under the circumstances. He has produced iron

oxides of unexpectedly high permanent magnet properties from a type of iron oxide not capable of developing such properties without the use of this unobvious (for the purpose) pre-treatment step. The sequence of the preliminary heating step and the subsequent reduction step is essential and is not in any way suggested by any prior art of record. This is the very essence of invention in chemical cases.

The rejection of the claims on the basis of claim 21 of the issued patent requires clarification. In making the rejection during the prosecution of the application, the Examiner contended that the claims of this application were *unpatentable over* the claimed subject matter of claim 21. In the decision on appeal, the Board states: "The rejection is as for double patenting". The question of double patenting had not been raised in connection with claim 21 prior to this appeal decision. Nor can such a proposition be logically presented under the facts of the case.

Claim 21 is directed to a method of making magnet material which comprises precipitating crystals of $Fe_2O_3 \cdot H_2O$ from solution of a particle size less than 6 microns in greatest dimension, heating said crystals in the air to a temperature between 500 and 1550° F. to drive off the water of hydration, and then reducing the dehydrated Fe_2O_3 crystals. This claim was based upon Example 3 of the patent. That example deals exclusively with particles of *acicular* particle shape and, indeed, any process for precipitating crystals of ferric oxide monohydrate necessarily and inherently results in the production of an acicular crystalline shape.

The claims on appeal specifically exclude the treatment of acicular type particles because the claims are limited to those materials which are beneficiated by the preliminary high temperature treatment to the extent of producing a coercive force value *that is higher than could be obtained by employing the same reducing conditions in the absence of the preliminary heating step*. As pointed out in the issued patent, the acicular ferric oxide monohydrate of Example I, when treated according to the reduction treatment therein described, achieves a coercive force value of 340 without any high temperature pre-treatment. That value

is obtained without the heading step to dehydrate the monohydrate that is
211 included in claim 21 of the patent.

By way of sharp contrast, the material produced as in Example I of the instant application, had a coercive force of 130 and a remanence of 500, so that it was substantially magnetic although not sufficiently magnetic to be a useful magnetic record medium. This material was of cubic particle shape as contrasted with the acicular particles of the examples of the patent. After treatment in air at 1300° F., followed by reduction at 600° F., the material achieved a coercive force value of 500 and a remanence of 250.

The criticality of the temperature range specified for the preheating step is evident from the results obtained in Example 5, which indicate that even with a pre-treatment at 700° F. (below the claimed range but sufficient to dehydrate ferric oxide monohydrate) followed by reduction, the powder still had a coercive force of only 150 and a remanence of 240.

Thus, the claims on appeal represent no extension of monopoly of the subject matter of claim 21. At the expiration of patent No. 2,694,656 the public at large would be free to produce acicular particles by precipitating crystals of $Fe_2O_3.H_2O$ from solution, dehydrate the crystals at a temperature between 500 and 1550° F., and reduce the dehydrated Fe_2O_3, all as described in claim 21. It is only with these types of iron oxides which do not in response to the reduction treatment acquire coercive force values in excess of 200 that the present claims are concerned. By their terms, the claims on appeal exclude specifically the materials which are described in the issued patent and defined in claim 21.

The same reasoning applies to appealed claim 39, which is directed to particles of cubic shape. Prior to applicant's present invention, there
212 was no known means for producing acceptable record members from iron oxide particles of cubic particle shape. Applicant's present process has provided a new source of raw material for the magnetic recording industry. While the specification of the issued patent mentions that forms other than the acicular crystalline form can be produced by

the process of the invention, there is no specific mention of the cubic particle shape. The fact that some of the claims of the applicant's issued patent may be broad enough to read upon shapes other than acicular does not amount to a specific disclosure of the cubic particle shape. Since there is no prior art whatever of record in the file of this application on the treatment of cubically shaped particles to render them suitable for magnetic record members, there is no basis in this record for the rejection of claim 39.

Reconsideration of the decision on appeal is requested in the light of the foregoing comments. A reversal of the previous decision is requested, or, in the alternative, a re-hearing of the appeal is solicited.

Respectfully submitted,
THE FIRM OF
/s/ Charles W. Hills
Attorneys for Applicant

Chicago 4, Illinois

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213 PLAINTIFF'S EXHIBIT NO. 1
(Paper No. 26)

[Mailed April 18, 1956]

DECISION OF THE BOARD OF APPEALS ON PETITION FOR RECONSIDERATION

A petition is presented requesting a rehearing of the appeal in this case or a reconsideration of our decision of March 8, 1956 wherein we affirmed that of the examiner in denying all of the claims on appeal.

Petitioner suggests we took a different view from that of the examiner in referring to "double patenting." The examiner held that the appealed claims did not distinguish from the claims of appellant's prior patent and he also held that the appealed claims were not patentable thereover in view of the prior art. In referring to "double patenting" we were directing attention to reliance on the claims of appellant's prior patent without reliance on unclaimed disclosure.

214 The quoted expression is sometimes used in a narrow sense in discussing a situation where precisely the same subject matter is claimed in different language. The term is also used in a broader sense to include situations where the pending claims are not addressed to the same subject matter as the patented claims but do not represent a patentable departure therefrom.

Petitioner suggests that we interpreted patented claim 13 in an unduly restricted manner by reading "providing a synthetic non-magnetic ferric oxide" as "demagnetizing ferric oxide". One method of "providing" such oxide is that recited in patented claim 21. Other methods are those of the Welo et al. citation, namely oxidizing and heating ferrosoferric oxide, and merely heating magnetic ferric oxide. Appellant is not in a position to now claim the method described in claim 21 of his patent. We are still of the opinion that the use of the Welo et al. method of "providing a nonmagnetic ferric oxide" is not a patentable departure from the subject matter of patented claim 13.

Petitioner also refers to the restriction to particles of cubic shape in claim 39. The discovery that the patented method, which is inclusive of the use of particles of cubic shape, is operative on particles of that shape, does not in our opinion warrant the grant of a second patent. Petitioner asserts that the patented process is not operative on particles of cubic shape, but it is not seen how he can support this assertion in the face of the terms used in the patented claims.

In view of the rights of other appellants waiting to be heard, this Board grants rehearings only under exceptional circumstances not here

215 presented. The request for rehearing is accordingly denied.

The petition has been considered but is denied with respect to making any change in our decision.

April 18, 1956

Charles W. Hills,
1414 Monadnock Block
Chicago 4, Ill.

/s/ E. W. Geniesse, Examiner-in-Chief
/s/ C. S. Duncombe, Examiner-in-Chief
/s/ I. Lisann, Examiner-in-Chief (Acting)
BOARD OF APPEALS

J.A. 93, R. 102
Defendant's Exhibit 1-A

Nov. 16, 1954 **M. CAMRAS** 2,694,656

MAGNETIC IMPULSE RECORD MEMBER, MAGNETIC MATERIAL
AND METHOD OF MAKING MAGNETIC MATERIAL
Filed July 25, 1947

Fig. 2

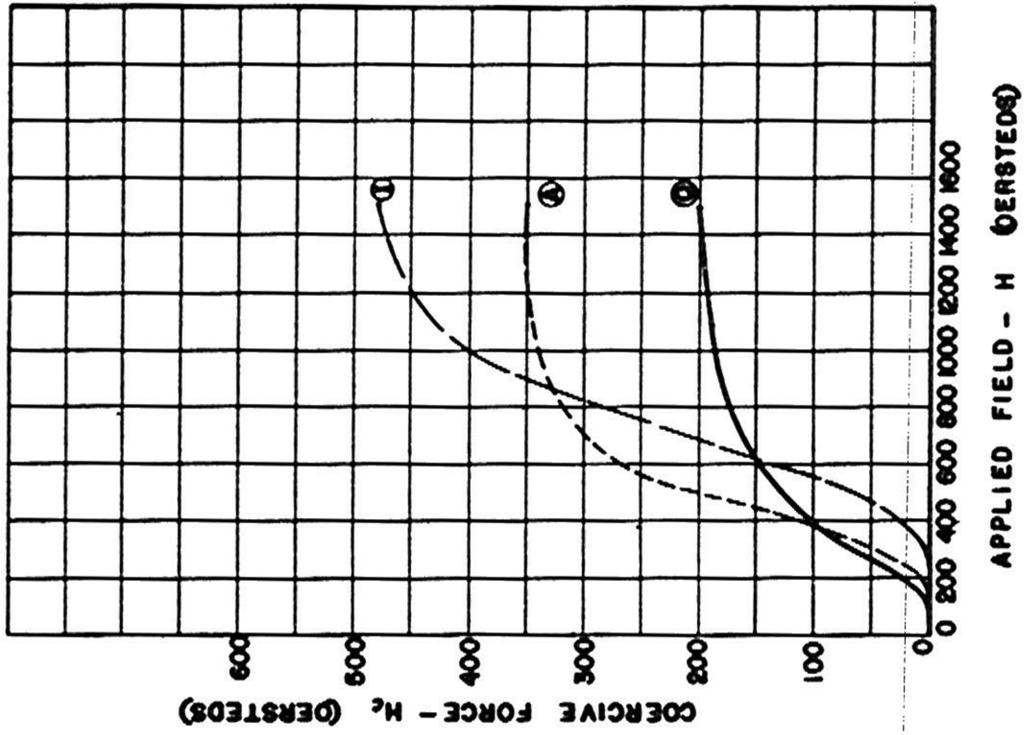
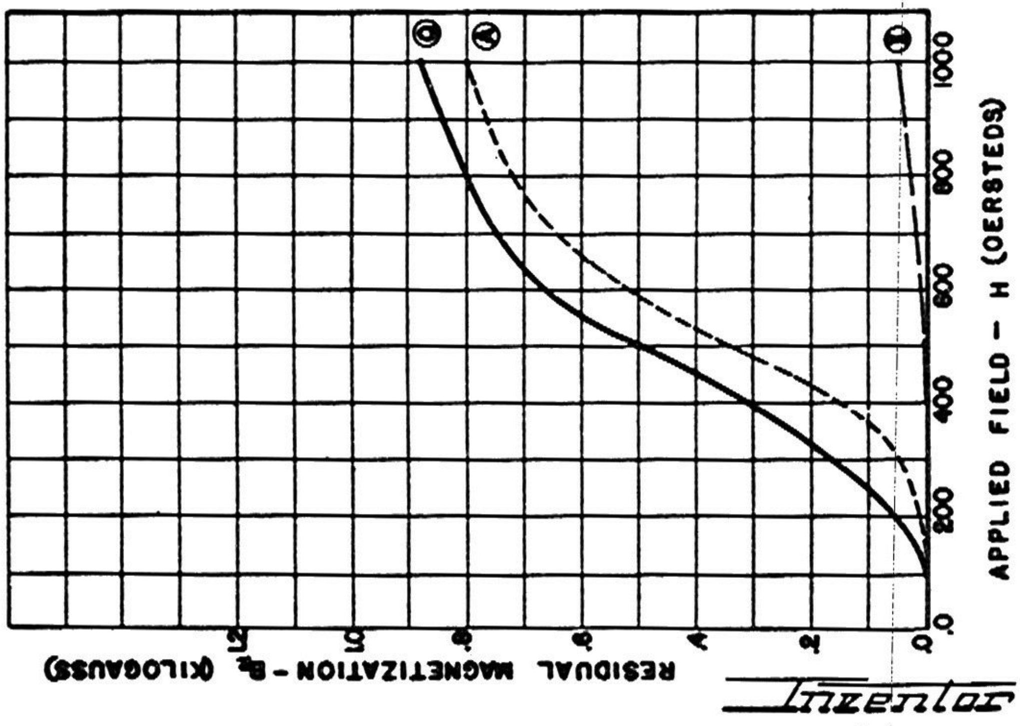


Fig. 1



Inventor
Marvin Camras

by The Firm of Charles W. Hills Attys.

J. A. 94, R. 103

United States Patent Office

2,694,656

Patented Nov. 16, 1954

2,694,656

MAGNETIC IMPULSE RECORD MEMBER, MAGNETIC MATERIAL, AND METHOD OF MAKING MAGNETIC MATERIAL

Marvin Camras, Chicago, Ill., assignor to Armour Research Foundation of Illinois Institute of Technology, Chicago, Ill., a corporation of Illinois

Application July 25, 1947, Serial No. 763,486

26 Claims. (Cl. 117—144)

This invention relates to permanent magnet material d to a method of making the same. More particularly, invention relates to the preparation of ferromagnetic aterial for use in a magnetic impulse record member comprising a base or carrier, such as a tape, ribbon or the like of non-magnetic material, coated or impregnated with a ferromagnetic track of my magnetic material.

Magnetic sound recorder tapes have heretofore been made with a non-magnetic backing, such as paper, plastic, or the like, and with a coating thereover, or an impregnation thereof, of a magnetic oxide of iron. Magnetic oxides of iron heretofore used for this purpose, however, have not been entirely satisfactory, since such magnetic oxides do not possess magnetic properties particularly suited for magnetic recording purposes. For one thing, prior magnetic oxides of iron have possessed relatively low coercive force values and relatively low energy products. They have also exhibited a rather high B_m to B_r ratio, usually considerably in excess of 3 to 1, whereas a relatively lower ratio gives better sensitivity and frequency response. B_m is expressed in gauss and is equal to maximum (B—H) and is termed the ferric induction. It is also equal to maximum $4\pi I$, where I is the magnetic intensity.

I have now found that it is possible to produce an iron oxide, or mixture of iron oxides, possessing magnetic properties that are much more desirable for magnetic impulse recording purposes. The magnetic material of my invention has a relatively high coercive force value, generally between 200 and 550 oersteds. This gives good high frequency response. My material also begins to saturate at fields as low as of from 400 to 600 gauss, a factor which makes for ease of erasing. In addition, my material has a B_m to B_r ratio of not over 3 to 1 and preferably considerably less, as compared with the higher than 3 to 1 ratio possessed by prior art magnetic iron oxides. This lower ratio gives better sensitivity and frequency response.

In addition, the magnetic material of my invention has high value of H_0, in that the B_r—H curve does not rise ppreciably until a field of about 250 gauss is reached. H_0 is the H value at the point where the B_r vs. H curve begins to rise rapidly. This magnetic characteristic reduces the tendency of the magnetic material to become affected by stray magnetic fields of relatively weak intensity, as for example the stray fields set up by closely arranged turns of the magnetized record member in the reels of a wire or tape recorder. Furthermore, my magnetic material exhibits a steep rise after reaching the H_0 value, a characteristic that makes for high recording sensitivity. The linear rise of the steep portion of the curve is responsible for a reduction in the distortion of the recording.

My magnetic material also exhibits a relatively high residual magnetism, B_r, which is a factor in improving low-frequency response and output. The combination of a relatively high coercive force, H_c, and a relatively high residual magnetism, B_r, together with the other desirable properties, has not been found to the best of my knowledge in magnetic iron oxides heretofore known. These, therefore, are properties that distinguish my material magnetically from prior art magnetic materials.

With regard to the physical characteristics of my magnetic material, I have found that in order to obtain the desirable magnetic properties above mentioned, it is preferable to use an iron oxide having a cubic lattice structure and an acicular crystalline structure. While ferric oxide, $Fe_2O_3.H_2O$, in acicular crystalline form is known, such ferric oxide is non-magnetic. I have now found, however, that this non-magnetic ferric oxide can be so treated as to render it magnetic, while still retaining its acicular crystalline form.

When treated in accordance with the method of my present invention, the non-magnetic ferric oxide is transformed into magnetic ferrosoferric oxide, Fe_3O_4, which may be used in that form as the magnetic material, or the magnetic ferrosoferric oxide so formed may be reoxidized back to a magnetic ferric oxide. In either case, the materials of my invention are preferably in acicular crystalline form, although other forms can be produced having, in general, similarly desirable magnetic properties. Apparently, the acicular crystalline form of the material is characteristic of the magnetic iron oxides possessing the best magnetic properties.

A further physical characteristic of my magnetic material is its small particle size. By proper control of the process, the crystals of ferric oxide used as the starting material are obtained in the form of uniformly small crystals having a length of the order of 1 to 2 microns. The magnetic material obtained by the further treatment of such ferric oxide in accordance with my present method is also in the form of crystals which in size, are of the same order of magnitude.

The small particle size of my magnetic material is conducive to better uniformity of sound reproduction and to a lower noise level. Furthermore, in the case of the acicular crystalline form of my material, there is a better opportunity of arranging the particles in overlapping relationship when used to coat or impregnate a paper tape, or the like. This, in turn, provides a more continuous and more uniform magnetic coating on the record member.

It is therefore an important object of this invention to provide novel permanent magnet material in the form of magnetic oxides of iron having preferably an acicular crystalline structure and being particularly adapted for use in the coating or impregnating of non-magnetic strip material to form a magnetic impulse record member of unusual efficacy.

It is a further important object of this invention to provide magnetic oxides of iron, including both ferric and ferrosoferric oxides and mixtures thereof, in the form of crystals of very small particle size and having magnetic properties superior to heretofore known magnetic oxides of iron.

It is a further important object of this invention to provide a method of preparing permanent magnet material from non-magnetic ferric oxides of acicular crystalline form to convert such non-magnetic oxides into magnetic oxides of iron having superior magnetic properties for use in the preparation of magnetic impulse record members.

It is a still further important object of this invention to provide a method of preparing magnetic material by the reduction of non-magnetic ferric oxide and to convert the same into magnetic ferrosoferric oxides, which may then be reoxidized to produce a magnetic ferric oxide, both the magnetic ferric and ferrosoferric oxides being in crystalline form of extremely fine particle size and having superior magnetic properties for use in magnetic sound recorders and the like.

Other and further important objects of this invention will be apparent from the disclosures in the specification and the accompanying drawings.

On the drawings:

Fig. 1 is a graph representing values of residual magnetism, B_r, expressed in kilogauss plotted against values of applied field, H, expressed in oersteds for several samples of magnetic material embodying my invention.

Fig. 2 is a graph of applied field vs. coercive force, H_c, for the same samples.

The starting material used in my method is preferably a hydrated ferric oxide, $Fe_2O_3.H_2O$, that exists in acicular crystalline form and which is available commercially. However, since I have found that it is desirable to prepare a fine, acicular crystalline form of ferric oxide especially for use in my method, the following example is given as a preferred method for its preparation:

EXAMPLE I

Three grams of caustic soda (NaOH) were dissolved in 10 grams of water. Twelve parts of copperas, $FeSO_4.7H_2O$, were separately dissolved in 60 grams of water. The caustic soda solution and the ferrous sulphate solution were then mixed in a vessel provided with agitation and the mixture agitated slowly for a number of hours, as for instance 17 hours, while constantly exposing a new surface of the mixture to the atmosphere. The reaction which took place yielded a thick greenish-yellow mass and may probably be expressed as follows:

$$(1)\quad 4FeSO_4.7H_2O+O_2+8NaOH\rightarrow 2Fe_2O_3.H_2O+4Na_2SO_4+30H_2O$$

The $Fe_2O_3.H_2O$ so produced is in colloidal form. The finely divided particles act as nuclei for growing larger crystals of $Fe_2O_3.H_2O$ in accordance with the subsequent steps about to be described.

In a separate vessel, 175 grams of $FeSO_4.7H_2O$ were mixed with 3.5 liters of water, and 1 kilogram of scrap iron was added. This mixture was then heated to 140° F., after which the thick greenish-yellow mass produced by Equation 1 was added and air bubbled through the resulting mass for about four hours, holding the same at about 140° F. The reactions that then occurred may be expressed as follows:

$$(2)\quad 4FeSO_4.7H_2O+O_2\rightarrow 2Fe_2O_3.H_2O+4H_2SO_4+22H_2O$$

$$(3)\quad H_2SO_4+Fe+7H_2O\rightarrow FeSO_4.7H_2O+H_2$$

The sulphuric acid formed in Equation 2 reacts with the scrap iron in accordance with Equation 3 to renew the amount of ferrous sulphate in solution. More iron may be added as the original iron dissolves.

The $Fe_2O_3.H_2O$ produced in accordance with the foregoing Equation 2 is obtained in the form of light yellow acicular crystals about ¼ to 1½ microns in length and about 1/10 to 3/10 micron in width. These size determinations were obtained by electron photomicrographs at 5000 magnification. A chemical analysis of the $Fe_2O_3.H_2O$ product showed 86% Fe_2O_3, and 12% H_2O.

The suspension of $Fe_2O_3.H_2O$ obtained as above described was filtered, washed, and dried at 212° F. This gave a filter cake that could be easily crushed to release the crystals in their original fine state of division. The material so produced was placed in a closed chamber, together with sharp-edged iron lumps to facilitate mixing. Hydrogen was passed through the chamber at a temperature in the chamber of 750° F., and the mixture tumbled until the powder had turned almost black. At this point, the amount of water vapor, which was heretofore being released in large volume, dropped off substantially, indicating that the reaction should then be stopped, since continued treatment beyond this point would result in deterioration of the material.

The material so prepared was then cooled by quenching the chamber with cold water while the gas was still passing through the chamber. Tumbling was continued until the product was at about room temperature before the gas supply was cut off. The chamber was then sealed and tumbled for about 10 minutes, with further cooling, before air was again admitted. If this precaution is not followed, the powder is apt to ignite.

The resulting powder is black and has an acicular crystalline structure. The particle size and shape are as previously described. The chemical analysis shows 78.7% Fe_3O_4 and 19.3% Fe_2O_3. However, the analysis may vary considerably. The extent of the reduction is indicated by the color of the material, which may vary from a dark brown to a deep black at room temperature.

EXAMPLE II

The product from Example I was mixed with a cellulose acetate binder and applied to a cellulose acetate base in the form of a tape, or ribbon, having a thickness of about 1.5 mils. As applied, the coating of the magnetic material had a thickness of about 1 mil, when dried. The coated strip was then run between calender rolls under a pressure of ten tons. This reduced the coating thickness to 0.5 mil and gave a smooth burnished surface. Magnetic measurements on the coated strip, identified as specimen A, gave the following properties:

Coercive force, $H_c=340$
Residual magnetization:
$B_r=420$ (before calendering)
$B_r=800$ (after calendering)
Maximum induction $-4\pi I$, $B_{fm}=640$ (before calendering)
Applied field, $H=1000$
Ratio $B_{fm}/B_r=1.6$

The crystals produced by Equation 2 of Example I were allowed to grow for the hereinafter specified various lengths of time instead of the 4 hours specified in Example I. Magnetic properties after processing in accordance with Example II were:

Specimen	No. Days of Crystal Growth	Color	H_c
B	½	light yellow	3
C	1	do	2.
D	2	yellow	275
E	5	orange yellow	230
F	8	dark orange yellow	220

The eight day crystals were about four times as large as the one day crystals. Preferred maximum dimensions of crystals are less than 1.5 microns and for a broader range less than 6 microns.

Although a temperature of 750° F. has been specified in the Example I for the temperature at which reduction by means of hydrogen is effected, the temperature may vary between a minimum of 500° F. and a maximum of about 1000° F. Between these two temperature limits, the times of treatment to effect reduction will depend upon the surface area of the particles, which, in turn, depends upon their fineness, the concentration of hydrogen in the reducing atmosphere and the concentration of water vapor in the reducing atmosphere. If water vapor or steam is mixed with the hydrogen in the reducing atmosphere, it helps to prevent over-baking, especially at the higher temperatures. Under-baked powder, indicated by its reddish color, has a low B_r. In fact, the B_r value may be too low to allow determination of magnetic properties if the powder is under-baked. In order to save hydrogen gas and also reduce the hazard, the reducing process can be started and ended with ordinary illuminating gas, switching over to a hydrogen atmosphere when the proper baking temperature of between 500 and 1000° F. is reached.

If the reducing reaction is carried on for too long a time or at too high a temperature, the coercive force decreases, the ratio of B_{fm}/B_r increases (which is undesirable), and the texture of the powder may be ruined due to the formation of lumps of large, hard particles. Ordinarily, if the temperature is kept within the limits of 500 to 1000° F., the time for the reaction to take place can be varied between ten minutes at the upper limit of temperature and 120 minutes or longer at the lower temperature limit.

In the following tables, as elsewhere herein, the magnetic properties were determined on coated strips, or tapes, prepared and calendered as described in Example II. Table I shows the variation of coercive force with reduction temperature:

Table I

Specimen	Temp., °F.	Time	H_c	Ratio: B_{fm}/B_r at $H=1{,}000$
G	300	16½ hours	(underbaked)	
H	400	120 min	do	
I	500	do	550	2.3
J	700	30 min	360	1.9
K	900	do	260	2.2
L	925	do	270	2.2
M	1,025	do	110	5.2
N	1,200	do	30	14.0

Table II indicates the variation of coercive values with the time of reduction:

Table II

Specimen	Temp., °F.	Time	H_c	Ratio: B_{tm}/B_r at H=1,000
P	700	30 min	300	1.9
Q	700	120 min	180	2.0
R	915	15 hours	140	5.1

The process of Example I may be varied in accordance with the following examples.

EXAMPLE III

Acicular crystals of $Fe_2O_3.H_2O$, as obtained by the reactions indicated by Equations 1 and 2 of Example I, were heated in air at temperatures between 500 and1550° F. to drive off the water of hydration. The crystalline structure was found to have changed from orthorhombic to hexagonal but with the crystalline form still acicular, while the color changed from light yellow to bright red. The particle size and shape were unchanged. The product so obtained had an analysis represented by the formula, Fe_2O_3.

The product so obtained was then subjected to the reducing and quenching steps described in connection with Example I. In general, the magnetic properties possessed by the final product of this example were approximately the same as those of the product produced by the process of Example I, although the coercive force, H_c, ran slightly higher.

This example indicates the feasibility of starting with the non-magnetic anhydrous form of ferric oxide, Fe_2O_3, rather than the hydrated form, $Fe_2O_3.H_2O$.

EXAMPLE IV

A product of Example I, identified as specimen S, and comprising a mixture of approximately 78.7% Fe_3O_4 and 19.3 Fe_2O_3, was placed in a shallow pan exposed to the air at a baking temperature of around 450° F. for ten hours or thereabouts. The particle size and shape of the product remained unchanged. The color was a light brown and the chemical analysis corresponded to that of Fe_2O_3. The magnetic properties of the product of this example were about the same as those of the previous examples, but with a higher B_r and a lower ratio of B_{tm}/B_r, as indicated by the following table:

Table III

Sample	Treatment	H_c	B_r	B_{tm}/B_r at H=1,000
S	before treatment	330	438	1.9
T	after reoxidation at 400° F	330	500	1.4

Reaction takes place at temperatures of about 300° F. or even lower, but oxidation is slower at the lower temperatures. In the temperature range of from 400 to 600° F. much of the powder is oxidized in relatively few minutes if good aeration is provided. The oxidation should be carried out at the lowest temperatures consistent with economical processing time. Higher temperatures progressively reduce the magnetic qualities. For example, the powder of sample S when treated at higher temperatures gave the following results:

Table IV

Sample	Treatment	H_c	B_r	B_{tm}/B_r at H=1,000
U	oxidized at 550° F	310	400	1.4
V	oxidized at 700° F	300	380	1.6

When the temperature is raised to about 1050° F. the magnetic properties fall off rapidly and are entirely lost at about 1200° F.

Either the product of Example I or that of Example III could be reoxidized by baking while exposed to the air, as in Example IV. The baking temperature of 450° F. is typical but the temperature may be varied between 300 and 900° F. The ferric oxide product so produced is largely gamma Fe_2O_3.

In Fig. 1 the residual magnetism, B_r, in kilogauss has been plotted against applied field, H, in oersteds for specimens A, I and Q. The full line curve for specimen Q and the dash line curve for specimen I indicate that these specimens have residual magnetization values that are within the limits for the magnetic material of my invention, although the B_r values for specimen I are on the low side. The dotted line curve for specimen A shows that the material represented by specimen A is of preferred magnetic qualities. It is desirable in order to prevent transfer that B_r at fields below 250 should be less than 20% of saturation and preferably under 15%. The B values at an applied field of 2000 oersteds have been considered to be the saturation values. As will be seen from an inspection of Figure 1, the B_r—H characteristic which is typical of the magnetic material of my invention, and of the sound tracks formed from such compacted magnetic material, rises most rapidly at fields between 200 and 600 oersteds and relatively slowly at fields between 0 and 200 oersteds and at fields above 600 oersteds. It is this magnetic characteristic that reduces the tendency of the magnetic material to become affected by stray magnetic fields of relatively weak intensity, such as the stray fields set up by closely arranged turns of the magnetized record member.

Fig. 2 represents coercive force, H_c, plotted against applied field, H, for the same samples and again indicates the superior magnetic properties of specimen A. It will be noted that all three specimens have a coercive force in excess of 200 at field saturation. The limits for H_c are from 200 to 550 and preferably from 275 to 400.

From the tables, it will be seen that these same samples A, I and Q have B_{tm} to B_r ratios at H=1000 of 1.6, 2.3 and 3.0, also within the upper limit of 3 to 1 previously mentioned.

Instead of effecting the reduction of $Fe_2O_3.H_2O$, or Fe_2O_3, by means of hydrogen gas, carbon monoxide gas may be used, or the ferric oxide may be reduced, by the use of other reducing agents, such as sulphur, sodium acetate, pyrogallol, and the like.

The following examples will serve to illustrate the use of these other reducing agents for reducing the ferric oxide used as starting material.

EXAMPLE V.—REDUCTION WITH PYROGALLOL

The $Fe_2O_3.H_2O$ of Example I was mixed with an excess of pyrogallol (about equal volumes of pyrogallol and ferric oxide) and heated gently to 400° F. for 15 minutes. The black residue was washed to remove excess pyrogallol. The dried material was then heated in the air to 700° F. to give a red material having a coercive force of 340 at a field of 2000 oersteds.

EXAMPLE VI.—REDUCTION WITH SULFUR

The $Fe_2O_3.H_2O$ of Example I was mixed with about 50% by weight of sulfur and the mixture heated to a temperature of about 800° F. Sulfur vapor formed above the mixture and excluded air. The black material formed as a result of the reaction was spread out into a thin layer and heated while exposed to air at a temperature of about 400° F. until it turned red. This yielded a material having an H_c of 210 at a field of 2000 oersteds.

EXAMPLE VII. — REDUCTION WITH SODIUM ACETATE

The $Fe_2O_3.H_2O$ of Example I was mixed with an excess of sodium acetate (dry) and heated for 3 hours at about 750° F. The black product was mixed with water, filtered and washed. The resulting black powder had a coercive force of 215 at a field of 1000 oersteds.

EXAMPLE VIII.—REDUCTION WITH HYDROGEN

Crystals of $FeSO_4.7H_2O$ were heated in an enclosed chamber to 750° F. for ½ hour in an atmosphere of hydrogen. The powder so formed was cooled, filtered and washed with water. The residue had a coercive force of 200 at a field of 1000 oersteds.

The magnetic products of any of the above described examples may be used for coating or impregnating purposes in the manufacture of a magnetic impulse record member.

As a summary of typical magnetic properties of the magnetic iron oxides of my invention, the following table is given.

Table V

Specimen	H_c at ($H=2,000$)	H_c at ($H=1,000$)	Sat. B_r[1] ($H=2,000$)	B_r ($H=1,000$)	B_r ($H=250$)	Ratio: $\frac{B_r(1,000)}{B_r(\text{Sat.})}$ percent	Ratio: $\frac{B_r(250)}{B_r(\text{Sat.})}$ percent	Ratio: $\frac{B_r(250)}{B_r(1,000)}$ percent	Ratio: $\frac{B_{rm}}{B_r}$ at $H=1,000$
A	370	340	912	800	14	87.7	1.54	1.75	1.9
I	540	400	75	45	<2	60.0	<2.67	<4.5	2.33
Q	215	185	1055	880	108	83.5	10.2	12.3	2.0
X	360	330	775	710	17.8	92.0	2.3	2.5	1.43
Y	325	310	525	500	15	95.0	2.86	3.0	1.65
W	280	245	530	500	100	94.0	18.7	20.0	1.66

[1] Considered saturated at $H=2,000$.

In the foregoing Table V, specimen X was prepared from a different initial batch of starting material in the same way as specimen A but was reduced at a somewhat lower temperature within the range of from 500 to 1000° F. Specimens Y and W were likewise prepared from different initial batches of starting material but otherwise were both prepared in the same way as specimen T.

It will be noted from the column headed "Ratio $B_r(1000)/B_r(\text{Sat.})$" that with the exception of specimen I, all specimens reported in the table show a ratio greater than 80%. Such a higher ratio is desirable for ease in magnetically erasing the record on the magnetic impulse record member.

Also, the column headed "Ratio $B_r(250)/B_r(\text{Sat.})$" shows that all of the specimens reported exhibited less than 20% of saturation value, and all but specimen W less than 15% of saturation value, at fields below 250. It is desirable that B_r at fields below 250 should be less than 20% and preferably less than 15% of saturation value in order to prevent the transfer of magnetism by reason of closely adjacent stray magnetic fields of relatively weak intensity. If my magnetic material did not posses this magnetic characteristic, portions of magnetized tape in a reel might magnetize adjacent unmagnetized turns of the tape and thus adversely affect the recording and reproducing fidelity of the tape.

The column headed "Ratio $B_{rm}/B_r(H=1000)$" shows that all of the specimens exhibited a ratio of 3 to 1 or less. This magnetic characteristic, as already explained, is important from the standpoint of better sensitivity and frequency response.

EXAMPLE IX

In this example, some of the magnetic Fe_2O_3 of specimen W, which had been produced by the method of Example IV followed by re-oxidation at 400° F., was re-reduced in accordance with the method of Example I. The coercive force was thereby increased from 260 to 405 at $H=2000$. This increase in coercive force indicates that the coercive force of a given sample of magnetic Fe_3O_4 or Fe_2O_3 can be improved by being repeatedly oxidized and reduced under the temperature conditions given herein.

The best prior art iron oxides that I have tested showed B_{rm}/B_r ratios at $H=1000$ of around 4, and H_c values at $H=1000$ of around 120. The superior magnetic properties of my magnetic iron oxide material are easily demonstrated by a comparison of the recording and reproducing performance of tapes made from such prior art iron oxides and my magnetic material.

I claim as my invention:

1. Ferromagnetic iron oxide adapted to form an element of a magnetic impulse record member, consisting essentially of uniformly small elongated crystals having characteristically in their as produced condition a length-to-width ratio of about 2.5 to 1 and higher of a synthetic ferrosoferric oxide, Fe_3O_4, formed from a non-magnetic iron oxide of the group consisting of alpha ferric oxide monohydrate and the anhydride thereof, said ferrosoferric oxide having a cubic crystal lattice and a coercive force, H_c, within the range of 290–360 oersteds.

2. Ferromagnetic iron oxide, adapted to form an element of a magnetic impulse record member consisting essentially of uniformly small elongated crystals having characteristically in their as produced condition a length-to-width ratio of about 2.5 to 1 and higher of a synthetic gamma ferric oxide, Fe_2O_3, formed from a non-magnetic iron oxide of the group consisting of alpha ferric oxide monohydrate and the anhydride thereof, said gamma ferric oxide having a cubic crystal lattice and a coercive force, H_c, within the range of 220–290 oersteds.

3. A ferromagnetic iron oxide material adapted to form an element of a magnetic impulse record member, said material consisting essentially of acicular crystalline particles uniformly small in size and not over 6 microns in their greatest dimension of a synthetic magnetic oxide of iron selected from the group consisting of magnetic ferrosoferric oxide, Fe_3O_4, and magnetic gamma ferric oxide, Fe_2O_3, the selected synthetic magnetic oxide of iron having a cubic lattice structure, and said material having a coercive force value of between 200 and 550 oersteds and a ratio of B_{rm}/B_r at $H=1000$ of not over 3 to 1.

4. A magnetic impulse record member having a non-magnetic carrier and a magnetic impulse track adherently bonded thereto of magnetic material and a binder therefor, said magnetic material consisting essentially of a magnetic synthetic iron oxide selected from the group consisting of ferrosoferric oxide, Fe_3O_4, and gamma ferric oxide, Fe_2O_3, said selected iron oxide being of a uniformly small particle size less than 6 microns in greatest dimension and having a coercive force of between 200 and 550 oersteds, said track having a rapid rate of rise in B_r for applied fields between about 200 and about 600 oersteds and a relatively slower rate of rise in B_r for applied fields below 200 and above 600 oersteds.

5. A magnetic impulse record member having a non-magnetic carrier and a coating adherently bonded thereto of magnetic material and a binder therefor, said magnetic material consisting essentially of a magnetic synthetic iron oxide selected from the group consisting of ferrosoferric oxide, Fe_3O_4, and gamma ferric oxide, Fe_2O_3, formed from a non-magnetic iron oxide of the group consisting of alpha ferric oxide monohydrate and the anhydride thereof, said selected iron oxide being in crystalline form of a uniformly small size less than 6 microns in greatest dimension and having a coercive force of between 200 and 550 oersteds, said magnetic material having a B_r versus H characteristic that rises most rapidly at fields between 200 and 600 oersteds and relatively slowly at fields between 0 and 200 oersteds and at fields above 600 oersteds.

6. As a new article of manufacture, a magnetic impulse record member comprising a thin, flexible, non-magnetic support, and adhered thereon, a layer of a magnetic synthetic iron oxide selected from the group consisting of ferrosoferric oxide, Fe_3O_4, and gamma ferric oxide, Fe_2O_3, formed from a non-magnetic iron oxide of the group consisting of alpha ferric oxide monohydrate and the anhydride thereof, said selected iron oxide in its as-produced condition being in the form of elongated particles having characteristically a length-to-width ratio of about 2.5 to 1 and higher and being of acicular crystalline form and of a uniformly small particle size less than six microns in greatest dimension and having a coercive force of between 200 and 550 oersteds.

7. The method of making permanent magnet material, which comprises precipitating a non-magnetic ferric oxide from solution in fine crystalline form, heating said non-magnetic ferric oxide in a reducing hydrogen atmosphere to a temperature of between 500 and 1000° F. for a sufficient length of time to reduce said ferric oxide to a magnetic ferrosoferric oxide having a coercive force of over 200 oersteds and a B_{rm}/B_r ($H=1000$) of not over 3 to 1.

8. The method of making permanent magnet material, which comprises precipitating a non-magnetic ferric oxide from solution in acicular crystalline form, heating said

non-magnetic ferric oxide in a reducing hydrogen atmosphere to a temperature of about 750° F. for a sufficient length of time to reduce said ferric oxide to a magnetic ferrosoferric oxide, cooling the ferrosoferric oxide in the presence of a reducing atmosphere to room temperature and then exposing said ferrosoferric oxide to the air to recover a ferrosoferric oxide having a coercive force value between 200 and 550.

9. The method of making magnetic material, which comprises precipitating a non-magnetic ferric oxide from solution in acicular crystalline form and of a particle size less than 6 microns in greatest dimension, heating said non-magnetic ferric oxide to a temperature between 500 and 1000° F. in a reducing atmosphere until the reduced oxide turns almost black, stopping the reaction at that point by cooling said reduced oxide in a reducing atmosphere to around room temperature and recovering ferrosoferric oxide of the same crystalline form and same order of particle size but having permanent magnet properties including a coercive force of over 200 oersteds and a $B_{tm}/B_r(H=1000)$ of not over 3 to 1.

10. The method of making magnetic material, which comprises precipitating a non-magnetic ferric oxide from solution in acicular crystalline form and of a particle size less than 6 microns in greatest dimension, heating said non-magnetic ferric oxide to a temperature between 500 and 1000° F. in a reducing atmosphere until the reduced oxide turns almost black, stopping the reaction at that point by cooling said reduced oxide in a reducing atmosphere to around room temperature and recovering ferrosoferric oxide of the same crystalline form and same order of particle size but having permanent magnet properties including a coercive force of over 200 oersteds and a B_{tm}/B_r $(H=1000)$ of not over 3 to 1 and reoxidizing said ferrosoferric oxide in the presence of oxygen at a temperature between 300° F. and 900° F. to produce a magnetic iron oxide consisting essentially of gamma Fe_2O_3.

11. The method of making permanent magnet material, which comprises precipitating a non-magnetic ferric oxide from solution in acicular crystalline form and of a particle size less than 6 microns, subjecting said non-magnetic ferric oxide to successive reducing, oxidizing, re-reducing and re-oxidizing steps to produce a magnetic material consisting essentially of gamma ferric oxide and having magnetic properties including a coercive force of over 200 oersteds and a B_{tm}/B_r $(H=1000)$ of not over 3 to 1, the reducing steps being carried out in a reducing hydrogen atmosphere at a temperature between 500 and 1000° F. and the oxidizing steps being carried out in the presence of oxygen at a temperature between 300 and 900° F.

12. The method of making permanent magnet material, which comprises growing acicular crystals of $Fe_2O_3.H_2O$ from solution of a particle size of less than 6 microns in greatest dimension, heating said crystals in the air to a temperature between 500 and 1550° F. to drive off the water of hydration, reducing said dehydrated crystals at a temperature between 500 and 1000° F. in a gaseous reducing atmosphere and cooling in a gaseous reducing atmosphere to obtain a magnetic material also in acicular crystalline form and of a particle size less than 6 microns and consisting essentially of Fe_3O_4 particles having a coercive force value of at least 200 oersteds and a B_{tm}/B_r $(H=1000)$ of not over 3 to 1.

13. The method of making magnetic iron oxide which comprises providing a synthetic non-magnetic ferric oxide in crystalline form and of a particle size not over 6 microns in its greatest dimension and reducing said non-magnetic oxide at elevated temperature to produce a ferrosoferric oxide having permanent magnet properties including a coercive force of at least 200 oersteds.

14. The method of making magnetic iron oxide which comprises providing a synthetic non-magnetic ferric oxide in acicular crystalline form and of particle size not over 6 microns in its greatest dimension, reducing said non-magnetic oxide at elevated temperatures to produce a ferrosoferric oxide and oxidizing said ferrosoferric oxide to gamma ferric oxide having permanent magnet properties including a coercive force of at least 200 oersteds.

15. The method of improving the magnetic properties of a synthetic gamma ferric oxide produced by the method of claim 14, which comprises reducing said gamma ferric oxide in a hydrogen atmosphere at about 750° F. and recovering an almost black iron oxide of higher coercive force than that of said gamma ferric oxide.

16. The method of making permanent magnet material, which comprises providing a synthetic non-magnetic ferric oxide in the form of fine elongated crystals of a length-to-width ratio of about 2.5 to 1 and greater and of less than 6 microns maximum dimension, heating said non-magnetic ferric oxide under reducing conditions at a sufficiently high temperature and for a sufficient length of time to reduce said ferric oxide to a ferrosoferric oxide also of elongated crystalline form but having permanent magnet properties including a coercive force of at least 200 oersteds.

17. The method as defined in claim 16, wherein the reduction is carried out in the presence of pyrogallol.

18. The method as defined in claim 16, wherein the reduction is carried out in the presence of sulphur.

19. The method of making permanent magnet material, which comprises providing a synthetic non-magnetic ferric oxide in the form of fine crystals of less than 6 microns maximum dimension, heating said non-magnetic ferric oxide in a reducing atmosphere of hydrogen at a sufficiently high temperature and for a sufficient length of time to reduce said ferric oxide to ferrosoferric oxide also in the form of fine crystals, stopping the reducing action at a point at which the said ferrosoferric oxide has a coercive force of over 200 oersteds and a B_{tm}/B_r $(H=1000)$ of not over 3 to 1, and recovering said ferrosoferric oxide having such magnetic properties.

20. The method of making permanent magnet material, which comprises subjecting material initially consisting essentially of a synthetic non-magnetic ferric oxide having a particle size not over 6 microns in maximum dimension to successive reducing, oxidizing, re-reducing and re-oxidizing steps to produce a magnetic material consisting essentially of gamma ferric oxide and having magnetic properties including a coercive force of over 200 oersteds and a B_{tm}/B_r $(H=1000)$ of not over 3 to 1, the reducing steps being carried out in a reducing hydrogen atmosphere at a temperature between 500 and 1000° F. and the oxidizing steps being carried out in the presence of oxygen at a temperature between 300 and 900° F.

21. The method of making permanent magnet material, which comprises precipitating crystals of $Fe_2O_3.H_2O$ from solution of a particle size of less than 6 microns in greatest dimension, heating said crystals in the air to a temperature between 500 and 1550° F. to drive off the water of hydration, reducing said dehydrated crystals at a temperature between 500 and 1000° F. in a gaseous reducing atmosphere and cooling in a gaseous reducing atmosphere to obtain a magnetic material also in crystalline form and of a particle size less than 6 microns and consisting essentially of Fe_3O_4 particles having a coercive force value of at least 200 oersteds and a B_{tm}/B_r $(H=1000)$ of not over 3 to 1.

22. The method of making magnetic material which comprises providing a synthetic non-magnetic ferric oxide in the form of fine crystals having a particle size of less than 1.5 microns in greatest dimension, heating said non-magnetic ferric oxide to a temperature between 500 and 1000° F. in a reducing atmosphere to form a ferrosoferric oxide, stopping said heating at a point where said ferrosoferric oxide has a coercive force of over 200 oersteds and a B_{tm}/B_r $(H=1000)$ of not over 3 to 1; cooling said ferrosoferric oxide in a reducing atmosphere to about room temperature, and recovering said ferrosoferric oxide in the form of fine crystals having a particle size of less than 1.5 microns in greatest dimension and having the aforesaid magnetic properties.

23. A method of making permanent magnet material, which comprises providing a synthetic non-magnetic ferric oxide in the form of fine elongated crystals of a length-to-width ratio of about 2.5 to 1 and greater and of less than 6 microns maximum dimension, heating said non-magnetic ferric oxide in a reducing atmosphere at a sufficiently high temperature and for a sufficient length of time to reduce said ferric oxide to a ferrosoferric oxide also of elongated crystalline form but having permanent magnet properties including a coercive force of at least 200 oersteds, and heating said ferrosoferric oxide exposed to the air at a sufficiently

high temperature and for a sufficient length of time to oxidize said ferrosoferric oxide to gamma ferric oxide having a coercive force of between 200 and 550 oersteds.

24. The method of making permanent magnet material, which comprises providing a synthetic non-magnetic ferric oxide in the form of fine elongated crystals of less than 6 microns maximum dimension, heating said non-magnetic ferric oxide in a reducing atmosphere selected from the group consisting of hydrogen, carbon monoxide and mixtures thereof at a sufficiently high temperature and for a sufficient length of time to reduce said ferric oxide to a ferrosoferric oxide; subjecting said ferrosoferric oxide in the presence of oxygen at a sufficiently high temperature and for a sufficient length of time to oxidize said ferrosoferric oxide to gamma ferric oxide having permanent magnet properties including a coercive force of at least 200 oersteds and recovering such gamma ferric oxide.

25. Ferromagnetic iron oxide selected from the group consisting of a synthetic ferrosoferric oxide, Fe_3O_4, and of a synthetic gamma ferric oxide, Fe_2O_3, adapted to form an element of a magnetic impulse record member, said iron oxide consisting essentially of uniformly small elongated crystals of less than about 1.5 microns maximum dimension having a length-to-width ratio of about 2.5 to 1 and higher, and having a cubic crystal lattice structure and a coercive force, H_c, within the range of 245 to 330 and remanence, B_r, of above about 500 gauss.

26. A magnetic impulse record member having a non-magnetic carrier and a coating adherently bonded thereto of a binder and magnetic material, said magnetic material being the ferromagnetic iron oxide defined in claim 25 and having a B_r versus H characteristic that rises most rapidly at fields between 200 and 600 oersteds and relatively slowly at fields between 0 and 200 oersteds and at fields above 600 oersteds.

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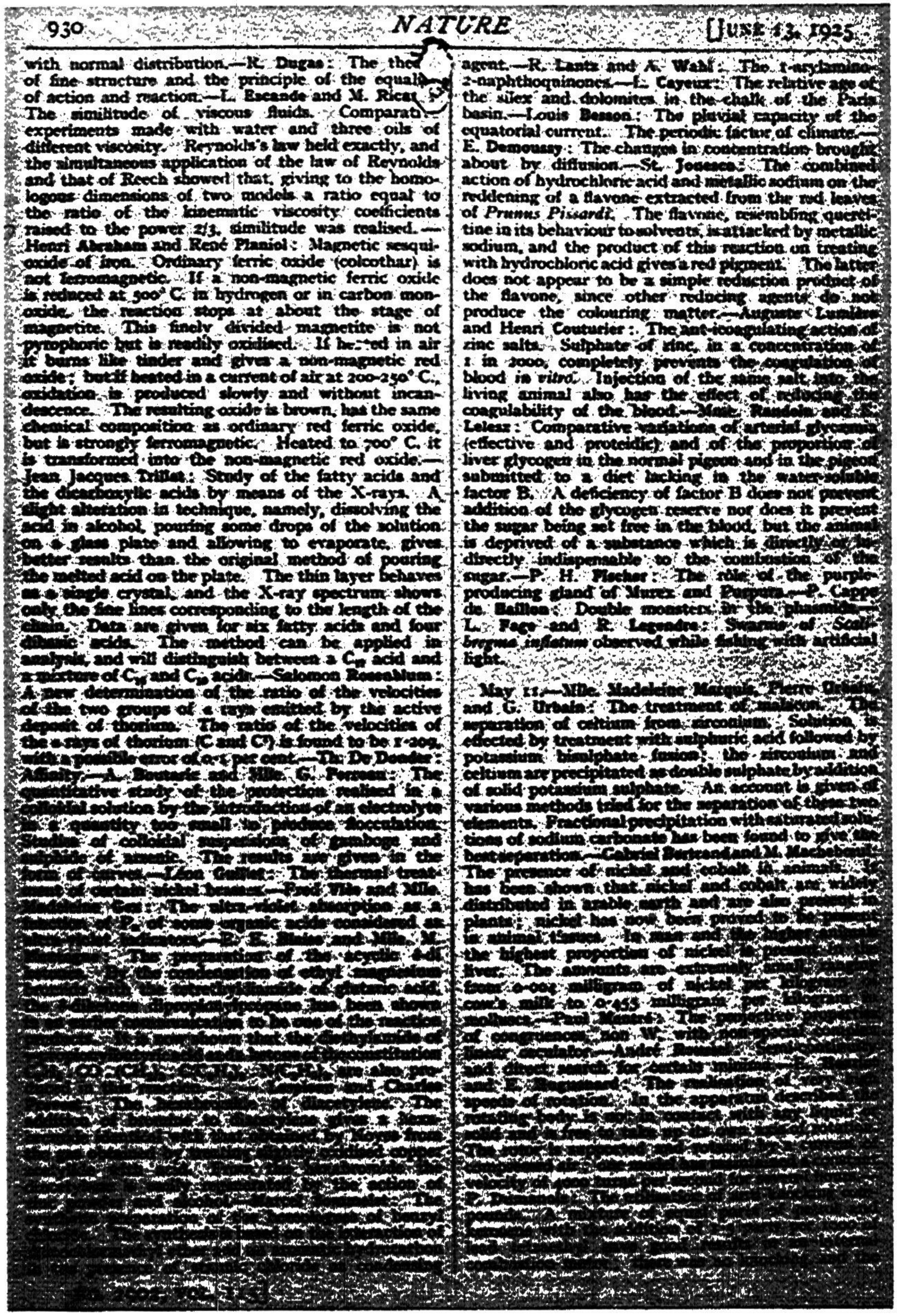
Defendant's Exhibit No. 1-B
[Abraham, et al. Article in
"Nature"]



with normal distribution.—**R. Dugas**: The theo[illegible] of fine structure and the principle of the equali[illegible] of action and reaction.—**L. Escande and M. Ricau**[illegible] The similitude of viscous fluids. Comparati[illegible] experiments made with water and three oils of different viscosity. Reynolds's law held exactly, and the simultaneous application of the law of Reynolds and that of Reech showed that, giving to the homologous dimensions of two models a ratio equal to the ratio of the kinematic viscosity coefficients raised to the power 2/3, similitude was realised.—**Henri Abraham and René Planiol**: Magnetic sesquioxide of iron. Ordinary ferric oxide (colcothar) is not ferromagnetic. If a non-magnetic ferric oxide is reduced at 500° C. in hydrogen or in carbon monoxide, the reaction stops at about the stage of magnetite. This finely divided magnetite is not pyrophoric but is readily oxidised. If heated in air it burns like tinder and gives a non-magnetic red oxide; but if heated in a current of air at 200-250° C., oxidation is produced slowly and without incandescence. The resulting oxide is brown, has the same chemical composition as ordinary red ferric oxide, but is strongly ferromagnetic. Heated to 700° C. it is transformed into the non-magnetic red oxide.—**Jean Jacques Trillat**: Study of the fatty acids and the dicarboxylic acids by means of the X-rays. A slight alteration in technique, namely, dissolving the acid in alcohol, pouring some drops of the solution on a glass plate and allowing to evaporate, gives better results than the original method of pouring the melted acid on the plate. The thin layer behaves as a single crystal, and the X-ray spectrum shows only the fine lines corresponding to the length of the chain. Data are given for six fatty acids and four dibasic acids. The method can be applied in analysis, and will distinguish between a C_{17} acid and a mixture of C_{16} and C_{18} acids.—**Salomon Rosenblum**: A new determination of the ratio of the velocities of the two groups of α rays emitted by the active deposit of thorium. The ratio of the velocities of the α-rays of thorium (C and C′) is found to be 1·209, with a possible error of 0·1 per cent.—**Th. De Donder**: Affinity.—**A. Boutaric and Mlle. G. Perreau**: The quantitative study of the protection realised in a colloidal solution by the introduction of an electrolyte in a quantity too small to produce flocculation. Studies of colloidal suspensions of gamboge and sulphide of arsenic. The results are given in the form of curves.—**Léon Guillet**: The thermal treatment of certain nickel brasses.—**Fred Vlès and Mlle. Madeleine Gex**: The ultra-violet absorption as a function of P of some organic acids considered as ultra-violet indicators.—**E. E. Blaise and Mlle. M.** [illegible]: The preparation of the acyclic [illegible] By the condensation of ethyl magnesium [illegible] [illegible]

agent.—**R. Lantz and A. Wahl**: The 1-arylamino-2-naphthoquinones.—**L. Cayeux**: The relative age of the silex and dolomites in the chalk of the Paris basin.—**Louis Besson**: The pluvial capacity of the equatorial current. The periodic factor of climate.—**E. Demoussy**: The changes in concentration brought about by diffusion.—**St. Jonesco**: The combined action of hydrochloric acid and metallic sodium on the reddening of a flavone extracted from the red leaves of *Prunus Pissardi*. The flavone, resembling quercitine in its behaviour to solvents, is attacked by metallic sodium, and the product of this reaction on treating with hydrochloric acid gives a red pigment. The latter does not appear to be a simple reduction product of the flavone, since other reducing agents do not produce the colouring matter.—**Auguste Lumière and Henri Couturier**: The anti-coagulating action of zinc salts. Sulphate of zinc, in a concentration of 1 in 2000, completely prevents the coagulation of blood *in vitro*. Injection of the same salt into the living animal also has the effect of reducing the coagulability of the blood.—**Mme. Randoin and E. Lelesz**: Comparative variations of arterial glycæmia (effective and proteidic) and of the proportion of liver glycogen in the normal pigeon and in the pigeon submitted to a diet lacking in the water-soluble factor B. A deficiency of factor B does not prevent addition of the glycogen reserve nor does it prevent the sugar being set free in the blood, but the animal is deprived of a substance which is directly or indirectly indispensable to the combustion of the sugar.—**P. H. Fischer**: The rôle of the purple-producing gland of Murex and Purpura.—**P. Cappe de Baillon**: Double monsters in the phasmids.—**L. Fage and R. Legendre**: Swarms of *Scolibregma inflatum* observed while fishing with artificial light.

May 11.—**Mlle. Madeleine Marquis, Pierre Urbain, and G. Urbain**: The treatment of malacon. The separation of celtium from zirconium. Solution is effected by treatment with sulphuric acid followed by potassium bisulphate fusion; the zirconium and celtium are precipitated as double sulphate by addition of solid potassium sulphate. An account is given of various methods tried for the separation of these two elements. Fractional precipitation with saturated solutions of sodium carbonate has been found to give the best separation.—**Gabriel Bertrand and M. Macheboeuf**: The presence of nickel and cobalt in animals. It has been shown that nickel and cobalt are widely distributed in arable earth and are also present in plants; nickel has now been proved to be present in animal tissues. In man and the higher animals the highest proportion of nickel is present in the liver. The amounts are extremely small, ranging from 0·004 milligram of nickel per kilogram in cow's milk to 0·455 milligram per [illegible]—**Paul Mentré**: The projective property of congruences non W. with [illegible]

Defendant's Exhibit No. 1-C
[Mellor, "Treatise on Inorganic and Theoretical Chemistry" Vol. 13]

Analyses were reported by H. A[illegible]laux,[2] J. J. Berzelius, W. Bornhardt, A. Breithaupt, A. Cathrein, A. H. Chester, F. W. Clarke, E. Classen, C. Desprets, E. Dittler and H. Hueber, J. N. von Fuchs, J. L. Gay-Lussac, F. F. Grout and T. M. Broderick, B. J. Harrington, M. F. Heddle, T. H. Holland, E. Hugel, C. J. B. Karsten, A. Knop, F. von Kobell, G. A. Koenig, A. Lacroix, S. M. Losanitsch, D. Lovisato, G. W. Maynard, J. A. Michaelson, W. G. Miller, T. Nordström, G. Nyiredy, T. Petersen, W. B. Phillips, F. J. Pope, C. F. Rammelsberg, L. Ricciardi, F. Scafile, B. Schwalbe, E. Sochting, J. del Pan, A. Stanojevic, J. Thiel, G. P. Tschernik, J. H. L. Vogt, T. L. Walker and A. L. Parsons, C. H. Warren, M. Weibull, and F. Zambonini. The results are, in the main, in agreement with the formula of *iron tritatetroxide*, or **ferrosic oxide**, Fe_3O_4, or $FeO.Fe_2O_3$, for the idealized mineral. The *ferropicotite* of A. Lacroix, found in Madagascar, has the composition $(Fe,Mg)O.(Al,Fe)_2O_3$. F. von Sandberger reported either stannous or stannic oxide in the magnetites of Hirschberg. Manganous oxide may be present, and M. Weibull writes the formula $(Fe,Mn)O.Fe_2O_3$; some magnesia and alumina were found by A. Knop to be associated with the mineral thus making the formula $(Fe,Mg)O.(Al,Fe)_2O_3$; and for the chromiferous varieties described by A. Cathrein, $FeO.(Fe,Cr)_2O_3$; while the titaniferous varieties may have $FeTiO_3$ or $MgTiO_3$ in solid soln. with Fe_2O_3; but, as shown by W. B. Phillips, magnetite exhibits a wide variation in composition, for the extremes in 30 analyses were:

	Fe as FeO	Fe as Fe_2O_3	$FeO : Fe_2O_3$
Magnetite (ideal) . . .	24·11	48·29	1 : 2·00
Octahedra from Vogelsberg . .	39·85	15·22	1 : 0·38
Shepard Mt., Missouri . . .	1·36	63·95	1 : 47·02

T. L. Walker found the dominant mineral in loadstone from Bon Accord, Transvaal, to be $(Fe,Ti)_2O_3$. J. D. Dana classed the magnetites as (1) ordinary massive, crystalline or sandy magnetites; (2) magnesian magnetites; (3) niccoliferous magnetites; (4) titaniferous magnetites; (5) manganesian magnetites; and (6) ochreous magnetites. W. N. Hartley and H. Ramage observed the presence of sodium, potassium, rubidium, silver, copper, gallium, manganese, lead, and nickel.

The existence of ferrosic oxide as a chemical individual was demonstrated by J. L. Gay Lussac, and J. J. Berzelius. J. L. Gay Lussac regarded it as a particular stage in the oxidation of iron, but J. Dalton, and J. J. Berzelius considered it to be a compound of ferrous and ferric oxides. The graphic formula is usually based on the assumption that the oxide contains both ferric and ferrous iron, as is exemplified by the presence of both salts of iron when ferrosic oxide is dissolved in acids. The graphic formula is $O{=}Fe{-}O{-}Fe{-}O{-}Fe{=}O$, or $Fe(FeO_2)_2$. S. H. Emmens, and W. G. Brown preferred formulæ in which the iron atoms were supposed to be quadrivalent. The formula $Fe(FeO_2)_2$ agrees with the spinel structure deduced by M. L. Huggins from the space-lattice.

According to S. Hilpert, the formation of the magnetic ferrosic oxide from non-magnetic, hydrated ferrous and ferric oxides shows that the magnetic property lies primarily in the combination of the two—where ferric oxide acts as an acid anhydride, and ferrous oxide as a base. This makes magnetic ferrosic oxide a ferrous ferrite, $Fe(FeO_2)_2$, *i.e.* a member of the family of spinels. That the magnetic property depends on the acidic property of the ferric oxide is shown by the fact that the ferrous oxide can be replaced by cupric, cobaltous, calcium, barium, and potassium oxide to form magnetic ferrites—*vide infra*. Indeed, cupric ferrite, $(CuO)^+(Fe_2O_3)^-$, is as strongly magnetic as $(FeO)^+(Fe_2O_3)^-$. It is possible to oxidize magnetite to magnetic ferric oxide; but in the compound $FeO.Fe_2O_3$, only the ferrous iron can be oxidized to furnish $(Fe_2O_3)^+(2Fe_2O_3)^-$, or *ferric orthoferrite*, $Fe'''(FeO_3)'''$, and this red powder is the true magnetic ferric oxide, *q.v.* The cobalt ferrite, $(CoO)^+(Fe_2O_3)^-$, can be similarly oxidized to $(Co_2O_3)^+(2Fe_2O_3)^-$. O. Mügge's study of the reduction products of hæmatite and the oxidation products of magnetite led him to suggest that magnetite is a solid soln. of ferric oxide in

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gave 3FeO.Fe_2O_3; and M. Beaujeu [illegible] C. Méne, who gave Fe_5O_9 and $Fe_{11}O_{12}$. E. J. Maumené gave 20FeO.9Fe_2O_3, i.e. $Fe_{38}O_{47}$, which is very near Fe_3O_4. H. Chandra, A. Kaufmann and F. Haber described a bluish-black oxide of the composition 2FeO.3Fe_2O_3, or Fe_8O_{11}, obtained by the action of potassium nitrate on a boiling soln. of ferrous sulphate rendered alkaline with ammonia; and H. Chandra, and O. Hauser obtained a 4FeO.Fe_2O_3 by dehydrating the hydrate (*q.v.*). E. J. Kohlmeyer also obtained indications of the formation of FeO.3Fe_2O_3; 3FeO.5Fe_2O_3; and 3FeO.4Fe_2O_3; and complex products were obtained by H. Wicht—*vide infra*, solid soln. of ferric oxide and ferrous oxide—but R. Ruer and M. Nakamoto, and A. Simon and T. Schmidt could not confirm this. H. St. C. Deville considered that the oxide FeO.Fe_2O_3, or rather Fe_4O_5, is formed when steam acts on iron at an elevated temp.; but P. P. Fedotieff and T. N. Petrenko showed that at 1000° to 1100°, the oxidation with steam or carbon dioxide proceeds $Fe \rightarrow FeO \rightarrow Fe_3O_4$; and with mixtures of steam and air, $Fe \rightarrow FeO \rightarrow Fe_3O_4 \rightarrow Fe_2O_3$. F. Wüst obtained 2FeO.$Fe_2O_3$ by heating ferric oxide in vacuo at 1100°; and 3FeO.Fe_2O_3, at 1200°. J. W. Döbereiner heated ferrous carbonate and obtained 3FeO.Fe_2O_3; E. Glasson, 4FeO.Fe_2O_3; and J. N. von Fuchs, A. Knop, and C. F. Rammelsberg, 5FeO.2Fe_2O_3; whilst from the oxalate, C. F. Rammelsberg obtained 4FeO.Fe_2O_3. The physical properties of these mixtures or solid soln. were examined by G. K. Burgess and P. D. Foote, E. Greulich, S. Hilpert and J. Beyer, R. B. Sosman and J. C. Hostetter, and H. Wicht. Analyses of mill scale were reported by A. Ledebur.

What was formerly called anhydrous *æthiops martialis*, or *l'oxyde de fer noir*, or *black oxide of iron*, approximates to ferrosic oxide, and was prepared by the incomplete oxidation of iron by air or steam. F. Küspert described a demonstration experiment for the combustion of iron in oxygen. Near the beginning of last century, ferrosic oxide was prepared by oxidizing iron in air by J. L. Gay Lussac, F. Stromeyer, and H. Davy. J. J. Berzelius showed that ferric oxide dissociates forming ferrosic oxide when it is heated, so that ferrosic oxide is formed when iron is heated in air at a high enough temp. even if ferric oxide were first produced. G. Ruer and E. Deiss, and A. Kropf said that the principal product obtained by burning iron in oxygen is ferric oxide; most others say ferrosic oxide. E. Mitscherlich obtained ferrosic oxide in octahedral crystals by burning iron in a blast-flame. W. G. Mixter, and W. A. Roth observed that the conversion of iron into ferrosic oxide is not complete when iron burns in oxygen, even at 12 to 15 atm. press. When the non-oxidized iron is removed by a magnet, W. G. Mixter obtained a product approximating Fe_3O_4. H. Wicht found that the fused product obtained by burning iron in oxygen has 2·5 to 10 FeO, Fe_2O_3; A. Mittasch's product with oxidizing gases under press. was Fe_3O_4; R. W. G. Wyckoff and E. D. Crittenden's product approximated Fe_3O_4; and E. Deiss and H. Leysaht added that with the slow combustion of iron in oxygen, ferric oxide is formed, and with rapid combustion, ferrosic oxide.

F. Bergius found that the reaction between iron and liquid water under press. furnishes ferrosic oxide in a form which is easily reduced; and E. V. Shannon regarded magnetite as a transient intermediate and unstable stage in the atmospheric oxidation of meteoric iron to limonite; and A. Payen, H. Ost, N. J. B. G. Guibourt, A. Ackermann, E. Deiss and G. Schikorr, and J. Huggett observed its formation in the earlier stages of the rusting of iron in aerated water. According to J. L. Gay Lussac, and C. Despretz, red-hot iron in contact with steam forms hydrogen and ferrosic oxide. Red-hot iron filings repeatedly sprinkled with water also yield the black oxide. Unlike C. N. A. de Haldat du Lys, H. V. Regnault found that the oxidation of heated iron by steam does not progress further than ferrosic oxide. J. A. N. Friend and co-workers found that when iron is heated in steam to [illegible] to [illegible], it acquires a film of magnetic oxide, and the layers underneath contain various proportions of ferrous oxide associated with the magnetic oxide in solid soln. If the iron is in thin sheets, the product, with a prolonged heating in steam, is all ferrosic oxide. S. Hilpert and J. Beyer observed that the

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converting iron into ferrosic oxide. [illegible] V. Regnault, and C. R. A. Wright and A. P. Luff obtained ferrosic oxide by the oxidation of hammer-slag, and roll-scale. It was also obtained by F. Wöhler by heating the hydrate, $Fe_3O_4.H_2O$, in a closed vessel, and by H. Moissan at a temp. of 300° in an indifferent gas. T. Sidot observed that octahedral crystals of ferrosic oxide are produced when ferric oxide is heated for a long time at a high temp., but not sufficient to fuse the oxide, and he added that the product exhibits magnetic polarity if the tube containing the oxide be placed in the earth's magnetic meridian. According to A. Simon and T. Schmidt, in the decomposition of ferric oxide at a constant press. of 10 mm., the isobar for the decomposition has a sharp break at 1300°, corresponding with the complete conversion of ferric to ferrosic oxide, without the formation of any solid soln., or intermediate oxide. The X-radiograms of ferric oxide were of an unknown type; those of ferrosic oxide, of the spinel type; and those of partially decomposed ferric oxide corresponded with a mixture of ferric and ferrosic oxides, and included no other lines. G. Rose obtained ferrosic oxide by heating ferric oxide with borax in a reducing flame; H. V. Thompson, by heating ferric oxide with an excess of sodium chloride for several hours; K. A. Hofmann and K. Höschele, by heating ferric oxide with magnesium chloride in a weak reducing atmosphere; F. Knapp, by heating 40 per cent. of ferric oxide with glass as a flux, and subsequently removing the glass by treatment with hydrofluoric acid; and E. Weinschenk, by strongly heating a mixture of ferric oxide, ammonium chloride and sulphur—if the temp. be not high enough, iron disulphide is produced. According to O. Mügge, and A. A. Read, when ferric oxide is maintained at 1500° in nitrogen, or, according to H. Moissan, if it be heated in the electric furnace, in air, it is converted into magnetite. H. Wicht said that the product obtained by fusion varies in composition between FeO and Fe_2O_3. H. Moissan thought that ferrosic oxide represents a definite stage in the reduction of ferric oxide or its hydrate at 350° to 400° in hydrogen, but, as shown in connection with ferrous oxide, the reduction is a continuous process. H. Moissan stated that if ferric oxide is heated to dull redness in hydrogen, saturated with water vapour at 90°, ferrosic oxide is formed. S. Hilpert and J. Beyer recommended heating the ferric oxide at 400° in hydrogen, saturated with water vapour at 50°, and so arranged that the gas and vapour passes over a heated copper spiral before it passes over the ferric oxide. H. Moissan heated, at 420°, ferric oxide in carbon monoxide and obtained ferrosic oxide; G. Lepetit preferred a 1 : 3-mixture of carbon monoxide and dioxide at 700° to 720° and at one to one-third atm. press. R. Schenck, and A. Laurent obtained ferrosic or ferrous oxide by a similar process. H. R. Gregory and G. MacDonald used producer gas; and K. Elbe, and S. Lilja, organic residues from distillation processes. L. de Launay obtained impure ferrosic oxide by heating in a sealed tube, at 250°, a mixture of ferric oxide and petroleum.

J. Durocher prepared ferrosic oxide by heating to redness a mixture of ammonium carbonate and ferrous chloride—some hæmatite is also formed; H. St. C. Deville and H. Caron, by melting iron fluoride and boric acid in a platinum crucible suspended above the fluoride, and all contained in a hermetically sealed carbon crucible; H. St. C. Deville, by passing hydrogen chloride over ferrous oxide heated at a high temp.; J. L. Gay Lussac, F. Stromeyer, A. Gautier, and E. A. Parnell, by passing steam over ferrous chloride at a dull red-heat; and A. Gorgeu, by heating molten ferrous chloride to bright redness in an incompletely closed crucible; J. von Liebig and F. Wöhler obtained the magnetic oxide as a heavy, black powder by fusing in air a mixture of ferrous chloride at a low red-heat with dry sodium carbonate and extracting the sodium carbonate with water. C. von Hauer obtained ferrosic oxide by heating to redness ammonium ferrous chloride, and also by passing a mixture of air and steam over heated ferrous chloride; E. Stirnemann, by the action of the vapour of ferric chloride and water on heated china clay, or a mixture of silica, sodium aluminate, and calcium carbonate; or, by heating in a bomb for 12 hrs., at 570° to 580°, a mixture of ferrous and ferric chlorides

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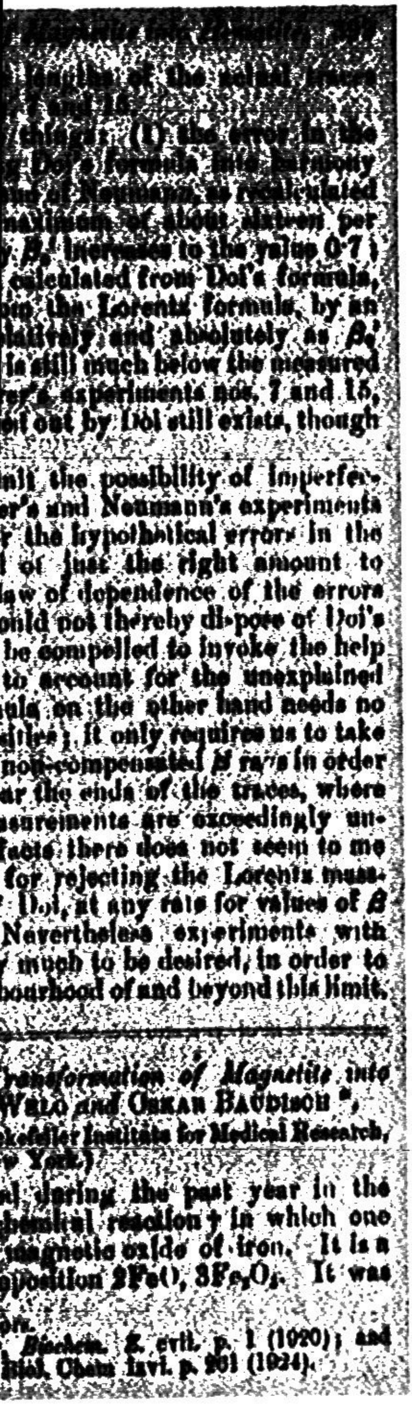
ferric oxide—the proportions being roughly as 1·3 : 1·0, and this excess seems to determine the more frequent formation of magnetite. Ferric pyrite, and ferrous pyrrhotite appear to follow the same rule of association. J. Morozewicz also observed that ferric oxide is formed only in silicates with a low proportion of ferrous iron, and is produced by melting acidic rather than basic silicates.

Spangles of what is possibly ferric oxide are frequently developed in pottery glazes highly charged with ferric oxide; and when deliberately produced, the products are called *aventurine glazes*, *sunstone glazes*, etc., though aventurine effects are produced in glazes and glasses in other ways. In the case of glazes highly charged with ferric oxide, the firing temp. and rate of cooling are important factors in producing the effects. The subject was discussed by H. Mäckler, P. Ebell, L. Petrik, etc. The formation of ferric oxide in molten silicates was also examined by F. Angel, C. Doelter, V. Hämmerle, G. Medanich, J. Morozewicz, K. Petrasch, H. H. Reiter, and J. H. L. Vogt. F. Kuhlmann crystallized the oxide from molten calcium chloride; and F. Parmentier, from molten potassium molybdate. H. B. Kosmann described a deposit of strongly magnetic limonite at Härteberg, Silesia; the hydrated ferric oxide contained no ferrous oxide, and he ascribed the ferromagnetic property to a "peculiar molecular grouping." Ferromagnetic hæmatite occurs as a light chocolate-brown powder in the gossan deposit at the Iron Mountain, California, and M. Sarzeau, and J. L. Smith described minerals which may have been ferromagnetic ferric oxide, but the evidence is not clear. P. A. Wagner observed it near Johannesberg, South Africa, and the deposit was described by G. Gilbert. P. A. Wagner proposed to call it **maghemite**. The term *oxymagnetite* was suggested by A. N. Winchell. G. Frebold and J. Hesemann believe that the ferromagnetic hæmatite is formed in nature only under a restricted supply of oxygen up to the temp. of 320°, whilst ordinary hæmatite is produced at a lower temp. J. Huggett observed that natural hæmatite and martites always contained some magnetic oxide.

J. Robbins described the preparation of a **ferromagnetic ferric oxide**, Fe_2O_3, by the oxidation of magnetite by heating it in air, or by fusing it with potassium nitrate. He showed that the product was free from ferrous iron, and that it could not in consequence owe its magnetic properties to the presence of ferrosic oxide, as is the case with ordinary magnetic hæmatite. The magnetic variety was also prepared by F. J. Malaguti by the oxidation with potassium chlorate of ferrous hydroxide and other hydrated iron oxide, and subsequent ignition. W. L. Dudley, G. Hauser, and A. Frenzel also obtained it by heating hydrated ferric oxide. A. Liversidge observed that some specimens of iron rust are ferromagnetic even when free from ferrous iron. This product was also obtained by E. F. Herroun and E. Wilson. R. Chevallier heated in air, at 350°, commercial, powdered, black magnetic oxide containing some free iron. H. Abraham and R. Planiol reduced ferric oxide with hydrogen or carbon monoxide at 500°, and obtained magnetite which on rapid re-oxidation at high temp. gives red, paramagnetic ferric oxide, but on slow oxidation in air at 200° to 250° gives yellowish-brown, strongly magnetic ferric oxide. R. B. Sosman and E. Posnjak prepared the ferromagnetic oxide by oxidizing precipitated ferrosic oxide, and also by dehydrating lepidocrocite, $Fe_2O_3.H_2O$, whilst they found that dehydrating goethite, $Fe_2O_3.H_2O$, gives only paramagnetic ferric oxide. J. Huggett obtained analogous results.

E. Wedekind and W. Albrecht concluded that the hydrated oxides are always less magnetic than the corresponding oxides; that in the range of 23 to 43 per cent. of water, the magnetic susceptibility decreases as the water of hydration increases; and that oxyhydrates, like artificial goethite, precipitated in the usual manner and heated in an autoclave, at various press., are ferromagnetic—vide hydrated ferric oxide. This subject was discussed by E. F. Herroun and E. Wilson, G. Chaudron and A. Girard, G. Gilbert, W. H. Newhouse and W. H. Callahan, P. A. Wagner, W. H. Newhouse, and L. H. Twenhofel. L. A. Welo and O. Baudisch obtained the ferromagnetic oxide by precipitating hydrated ferric oxide from a soln.



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ransformation of *Magnetite* into
Welo *and* Oskar Baudisch *,
kefeller Institute for Medical Research,
w York.)

d during the past year in the
hemical reaction † in which one
magnetic oxide of iron. It is a
position $2FeO, 3Fe_2O_3$. It was

on.
Biochem. Z. cvii. p. 1 (1920); and
Biol. Chem. lxvi. p. 201 (1924).

J. A. 105, R. 114

Defendant's Exhibit No. 1-D

[Welo and Baudisch, "Two Stage Transformation of Magnetite into Hematite"]

J.A. 106, R. 115 and 116

natural to compare its properties, both physical and chemical, with those of the more common ferro-ferric oxide FeO, Fe_2O_3, which is magnetite and usually written Fe_3O_4. At the same time a comparison was made with the oxide of Haber and Kaufmann*, which also has the composition $2FeO$, $3Fe_2O_3$, but is made in a different manner. During this work of comparison it was discovered that the process of transformation of each of these ferro-ferric oxides into the non-magnetic ferric oxide Fe_2O_3 takes place in distinct stages, and that the disappearance of the magnetism by heating is dependent on the existence of a preliminary oxidation.

Our work on the intercomparison of the properties of the three oxides is about complete, and a full report on it is to be published. In the present paper we shall be concerned only with the conditions which govern the transformation of each of these magnetic ferro-ferric oxides into the non-magnetic ferric oxides. The conditions are the same for each of them; at least they are qualitatively so. It will be sufficient, for the purpose of this paper, to describe the steps of the transformation of the magnetite, FeO, Fe_2O_3, into the non-magnetic, purely ferric oxide, Fe_2O_3.

Preparation of Magnetite.

The magnetite used in this study was the black, microcrystalline substance formed when a solution containing one mol of ferrous sulphate and two mols of ferric sulphate is poured into a boiling solution of sodium hydroxide. It is necessary that sodium hydroxide be present in excess so as to give, always, an alkaline solution. The precipitate was washed until the filtrate showed no alkali when tested with litmus paper. After collection on a filter the oxide was dried in a desiccator.

The Qualitative Observations.

Generally speaking, the facts observed on heating the oxide in a stream of oxygen are as follows:—The colour of the oxide changes suddenly from black to a dark red at about 220° C.; the oxide remains magnetic, although the iron has been completely converted into the ferric form so that the composition has become Fe_2O_3. The ferricyanide test showed not a trace of Prussian blue, and hence no

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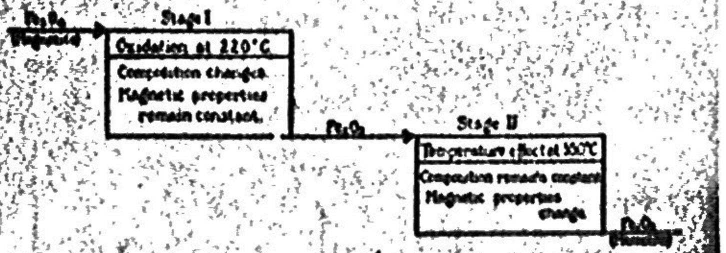
* F. Haber and A. Kaufmann, *Z. Elektrochem.* vii. p. 733 (1900–1901).

ferrous iron after oxidation for a sufficient time at 220° C. A solution of the oxide in hydrochloric acid titrated with potassium permanganate likewise yielded no iron in the divalent form. All of this shows that Fe_3O_4 may be completely oxidized to Fe_2O_3 at a temperature of 220° C. without loss of the magnetic properties.

On further heating, the oxide, now Fe_2O_3, remains magnetic until a temperature of 550° C. is reached. At this temperature a non-reversible transformation takes place, for the oxide becomes permanently non-magnetic. This transformation is obviously a pure temperature effect, since the oxidation was already complete at the lower temperature of 220° C. But to test the point a sample which had first been oxidized at 220° C. was heated in an atmosphere of nitrogen carefully freed from oxygen. It again became non-magnetic. The necessary condition, however, for the loss of magnetism at 550° C. is that the Fe_3O_4 must first be oxidized to Fe_2O_3 at some lower temperature. If Fe_3O_4 is heated in nitrogen without a preliminary oxidation it is stable, and retains its magnetic properties even if heated to 800° C. These observations are briefly summarized in the scheme shown in fig. 1.

Fig. 1.


Fe_3O_4
Magnetite
Stage I
Oxidation at 220°C.
Composition changes.
Magnetic properties remain constant.
Fe_2O_3
Stage II
Temperature effect at 550°C
Composition remains constant
Magnetic properties change
Fe_2O_3

It has been stated that the oxidation begins at 220° C. A more precise statement would be that the oxidation becomes sufficiently rapid at that temperature so that the change in colour may be observed. It is recognized that oxidation may take place at a much lower temperature.

It is interesting to note here that the temperature of stage II. is found to be nearly the same as, and probably does actually coincide with, the Curie point for natural magnetite, which is 525° C., according to Graetz*.

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* Graetz, 'Handbuch der Elektrizität und des Magnetismus,' vol. iv. p. 888.

The Permeabilities.

It was decided to make more precise measurements of the permeabilities on the original Fe_3O_4 and on the derived oxides after each stage of the transformation. A long magnetising solenoid of two layers was wound on a brass tube. There were 13·6 turns per centimetre, so that the field intensity turns out to be 17·1 gauss per ampere. Another brass tube was made to fit into the tube carrying the solenoid. On this second brass tube circular grooves were cut to make room for test coils of, respectively, 4, 20, and 100 turns. The leads were carried to binding-posts at one end so that any one of the coils could be connected to a high sensitivity Leeds & Northrup ballistic galvanometer. The oxides were packed by jarring (not by tamping) into a glass tube which slipped into the second brass tube. The length of packing was always 20 centimetres, so the demagnetizing factor becomes entirely negligible. The densities of packing could not be regulated, but were never far from an average value of 1·32 $\frac{gr.}{cm.^3}$. The corrected permeabilities μ_c were calculated from the observed permeabilities μ_o by the relation

$$\mu_c = (\mu_o - 1)\frac{1\cdot 32}{\delta_p} + 1,$$

where δ_p is the density of packing.

The fact that these oxides have permeabilities of about 3 at the most simplifies the procedure. The galvanometer deflexions with oxide in the glass tube and with the tube empty are of the same order of magnitude, and their ratio R, for a given magnetizing field, can be introduced directly into the formulas.

Let A_o be the internal cross-section of the glass tube containing the oxide, and A_t be the mean area of the test-coil wound on the brass tube.

With the empty tube the flux through the coil is A_tH and the deflexion

$$d_e = CA_tH,$$

C being a constant.

With oxide in the tube the flux is

$$(A_t - A_o)H + A_oB = (A_t - A_o)H + A_o\mu H,$$

so that the deflexion

$$d_o = C[(A_t - A_o)H + A_o\mu H],$$

$$\frac{d_o}{d_e} = \frac{(A_t - A_o)H + A_o\mu H}{A_tH} = R,$$

giving

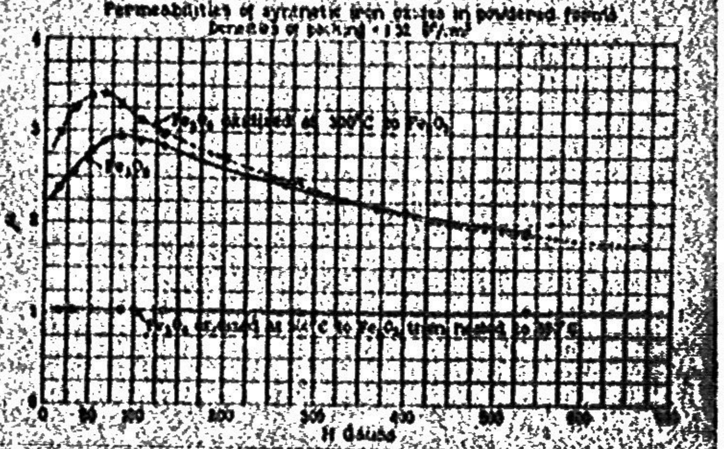
$$\mu = \frac{A_t}{A_o}R - \frac{A_t - A_o}{A_o},$$

and, if we desire, an equally simple derivation of the induction

$$B = \frac{H}{A_o}[RA_t - (A_t - A_o)].$$

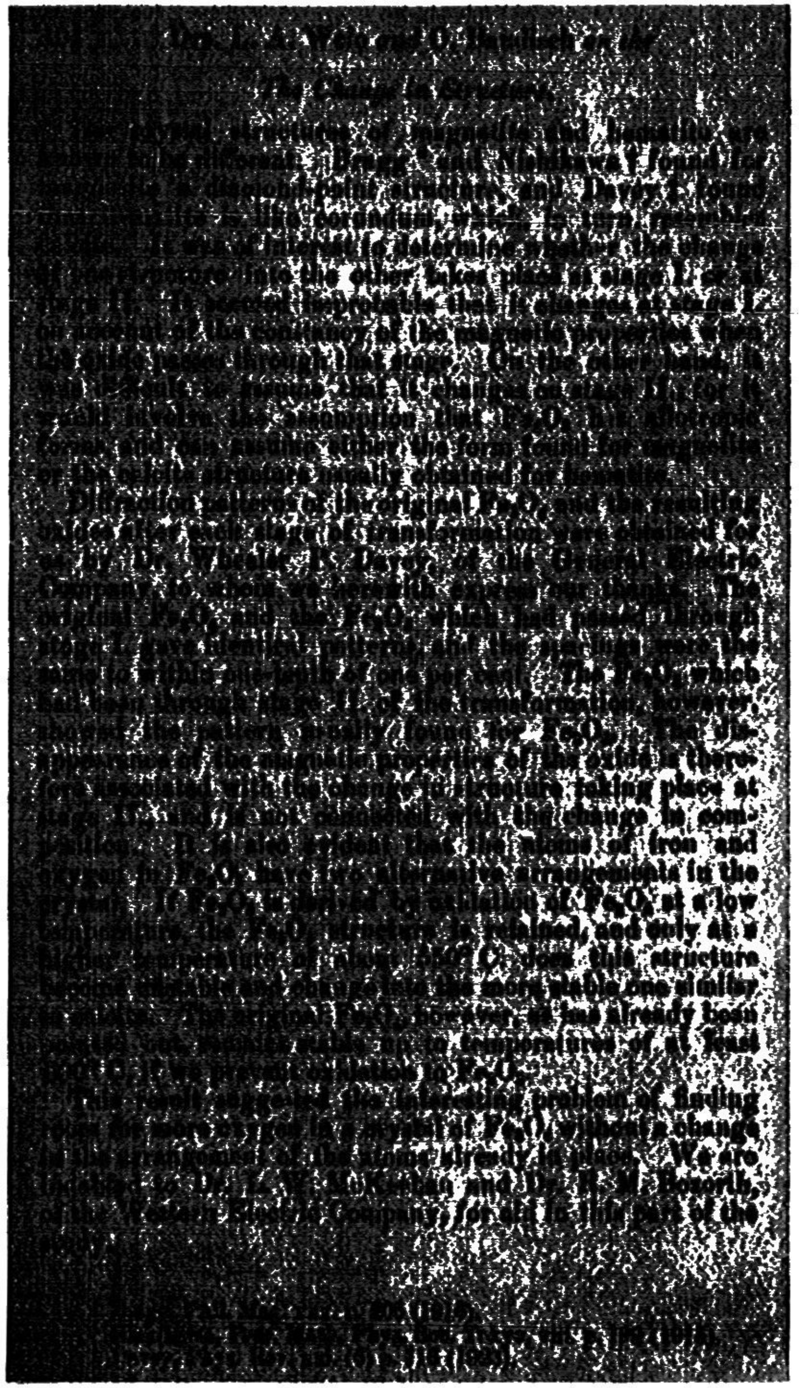
Field intensities greater than 85 gauss could not be obtained without undue heating of the solenoid, so that another was used for the stronger fields. It carried several hundred turns. As it was rather irregularly wound it had to be calibrated, and was found to give a field intensity of 13·2 gauss per ampere.

Fig. 2.



The results of the magnetic measurements are shown in fig. 2. Our qualitative observations are confirmed. The maximum permeability of the oxide after having passed the first stage of transformation is somewhat higher than that of the original Fe_3O_4, and there is shown a considerable shift of its position. No reason is known for this. At higher fields the curves coincide, which accounts for our qualitative observation (made with high field intensities) that the magnetic properties remain unchanged on oxidation at a low temperature. After the second stage the permeability is seen to be nearly, but not quite, unity at all fields. The maximum residual permeability is 1·015, and is so small that the material would be considered non-magnetic towards rough qualitative tests.

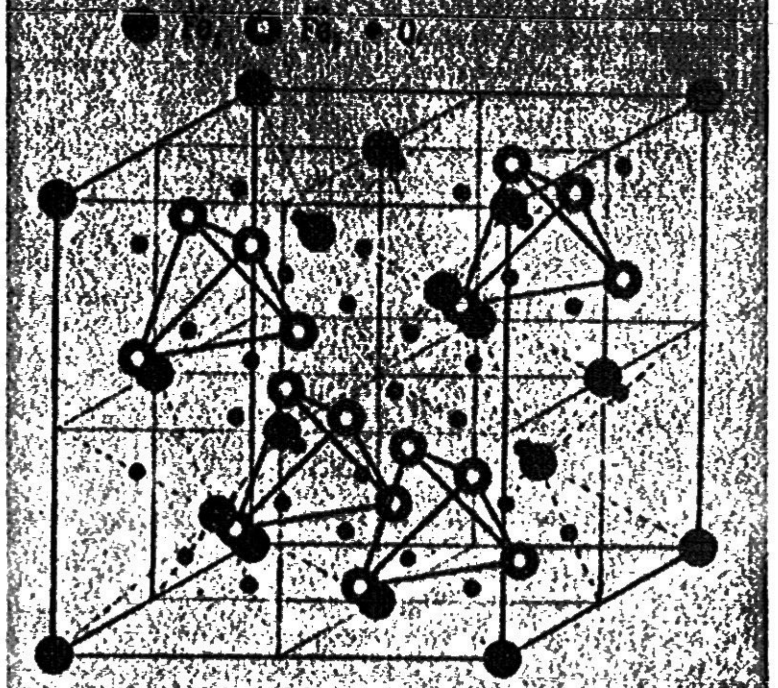
J.A. 108, R. 119, 120



Transformation of Mag[illegible]e into Hematite.

Fig. 4 shows the unit cube for magnetite as constructed from data given by Wyckoff *. The unit cube contains eight molecules, so that Fe_3O_4 may be written [illegible]

Fig. 4.



MAGNETITE

trivalent iron atoms, and they are thus shown in fig. 4. In view of the uncertainties as to the actual radii of combination, it is quite possible for the extra oxygen atoms to be placed, as shown, without undue distortion of the system. The edg of the unit cube has a length $a = 8{\cdot}30$ Å., and the distan from the centre of the tetrahedron formed by trivalent ir atoms to each corner is

$$\frac{a}{8}\sqrt{3} = 1{\cdot}795 \text{ Å}.$$

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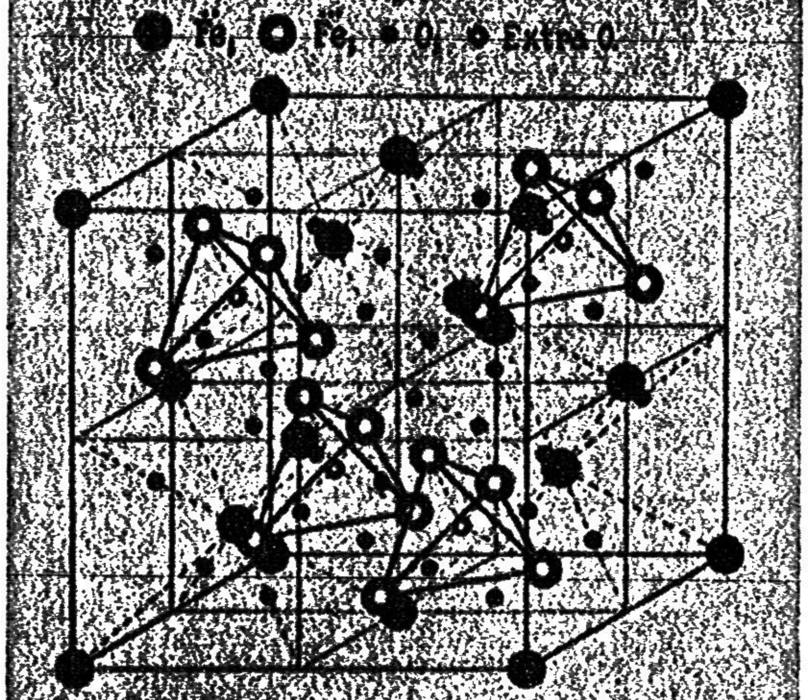
* Wyckoff, Journ. Franklin Inst. ccxv. pp. 531, 544 (1923).

The radius of the oxygen atom is usually taken to be 0·65 A. This leaves 1·145 A. as a remainder for the radius of the iron atom. The radius of the iron atom appears to be variable, and values as low as 1·23 A. have been found in the case of pyrites * and in iron itself.

An alternative and perhaps equivalent arrangement would be to place an oxygen atom at the middle of each edge of the unit cube. The room at our disposal would be the same. This question is not considered here, as it would require a more detailed study of the diffraction pattern, and would probably be indeterminate even then. We are here concerned only with the possibility of putting more oxygen in magnetite so as to change it to hematite without changing the existing arrangement of the atoms.

Fig. 1.



OXIDIZED MAGNETITE

A few remarks may appropriately be added here regarding an earlier objection to our interpretation which has now no force, since we see that it is possible to convert Fe_3O_4 to

Fe_2O_3 without disturbing the structure. The assumption was made that on the first stage the oxidation was only superficial, and that the cores of the particles remained unchanged. It was supposed that complete oxidation took place only at the higher temperature. The superficial layer of Fe_2O_3 formed, then, on stage I. would form so small a proportion of the whole that the magnetic properties and the diffraction pattern would appear unchanged. The fact that the oxide which has been through stage I. loses its magnetism and changes its structure when heated in nitrogen to 550° C. makes this improbable. The improbability is heightened by the fact that unoxidized Fe_3O_4 remains magnetic after heating in nitrogen to a still higher temperature. The nitrogen cannot contain sufficient oxygen as an impurity to bring about the necessary oxidation. That the oxidation is not superficial is shown also by the chemical tests for ferrous iron, for in these tests the whole particle, core and surface, is first dissolved in hydrochloric acid. To make doubly sure, a test was made for increase in weight when the transformation at stage II. takes place in a stream of oxygen. Instead of the calculated increase of 32 milligrams for the particular sample used, there was a decrease in weight of about one milligram.

The Order of the Oxidation Process.

We are now in possession of an unforced explanation as to why the presence or absence of magnetism in the synthetic ferric oxide Fe_2O_3 depends on the order in which the chemical operations are carried out. Hilpert * oxidized an existing ferro-ferric oxide into the ferric oxide by means of the strong oxidizing agents H_2O_2 and $K_2S_2O_8$, and found the product to be magnetic. On the other hand, we † have precipitated ferrous hydroxide in the presence of the same oxidizing agents, and the resulting oxide Fe_2O_3 is non-magnetic. The two cases may be represented by

$$(1)\ FeSO_4 + 2NaOH + xO_2 \rightarrow xFe_3O_4 + x\begin{bmatrix} H_2O_2 \\ \text{or} \\ K_2S_2O_8 \end{bmatrix} \rightarrow \underset{\text{Magnetic.}}{xFe_2O_3}$$

$$(2)\ FeSO_4 + 2NaOH + x\left\{\begin{matrix} H_2O_2 \\ \text{or} \\ K_2S_2O_8 \end{matrix}\right\} \rightarrow \underset{\text{Non-magnetic.}}{xFe_2O_3}$$

In case (1) the oxide first formed is magnetite, and it retains its structure in spite of the subsequent oxidation to Fe_2O_3. In case (2) the conditions of precipitation are such that the usual Fe_2O_3 structure is built up from the start.

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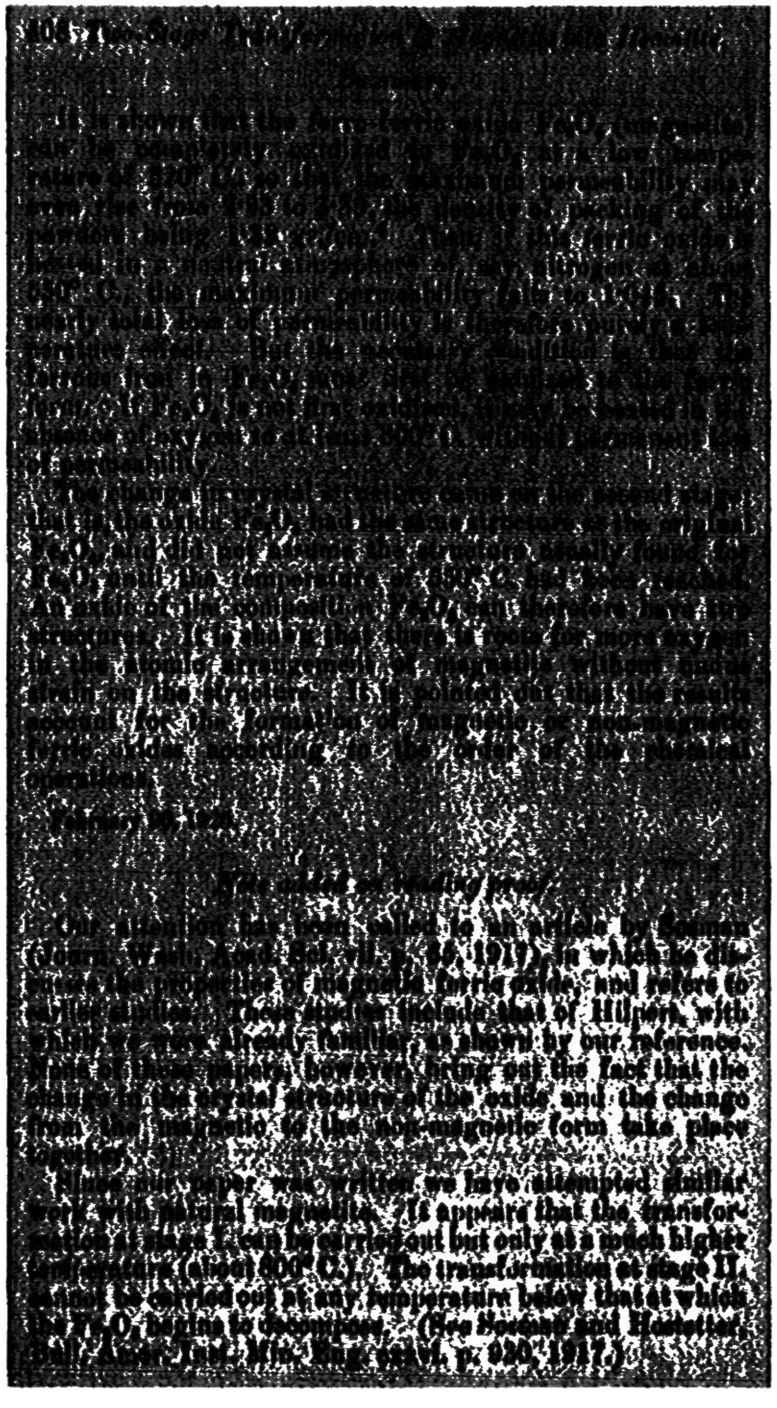
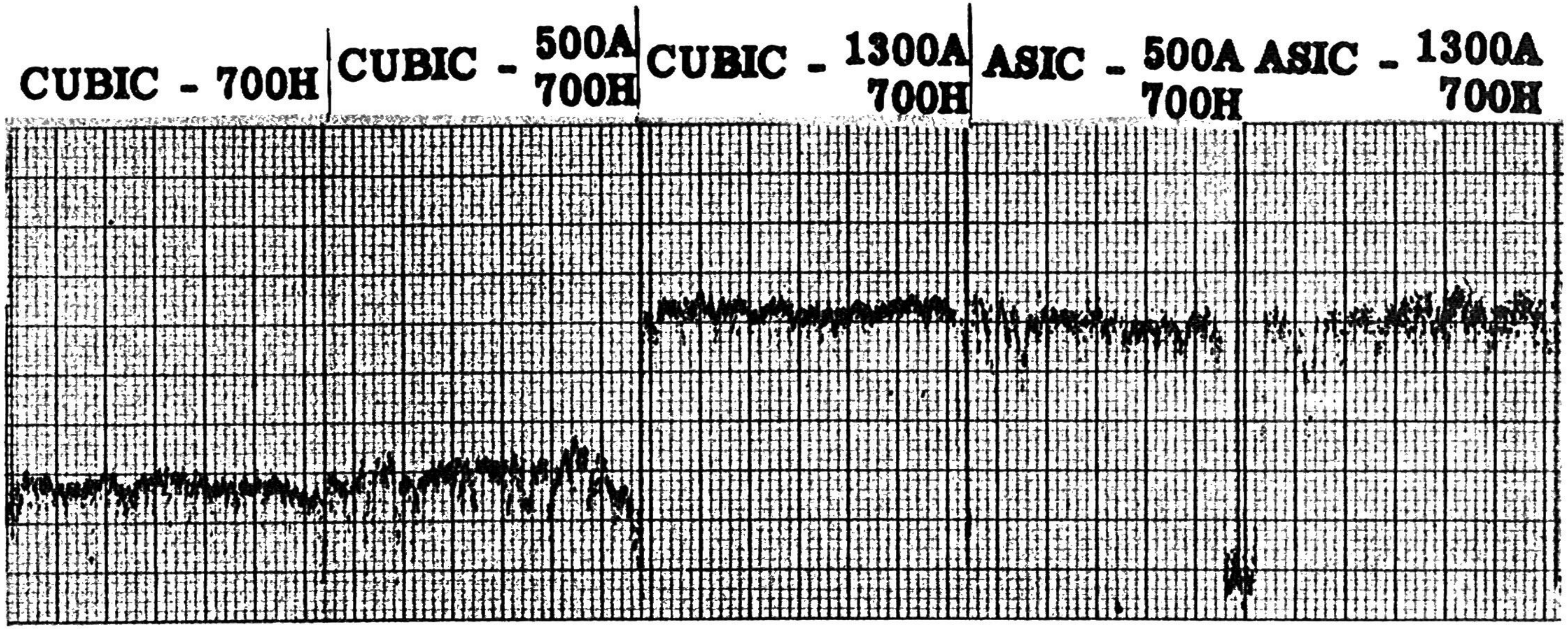
* 'X-Rays and Crystal Structure,' W. H. and W. L. Bragg, 4th Edition, p. 172.

* Hilpert, *Ber. Chem. Ges.* xlii. p. 2248 (1909).

† O. Baudisch and L. A. Welo, Journ. Biol. Chem. lxi. p. 261 (1924).

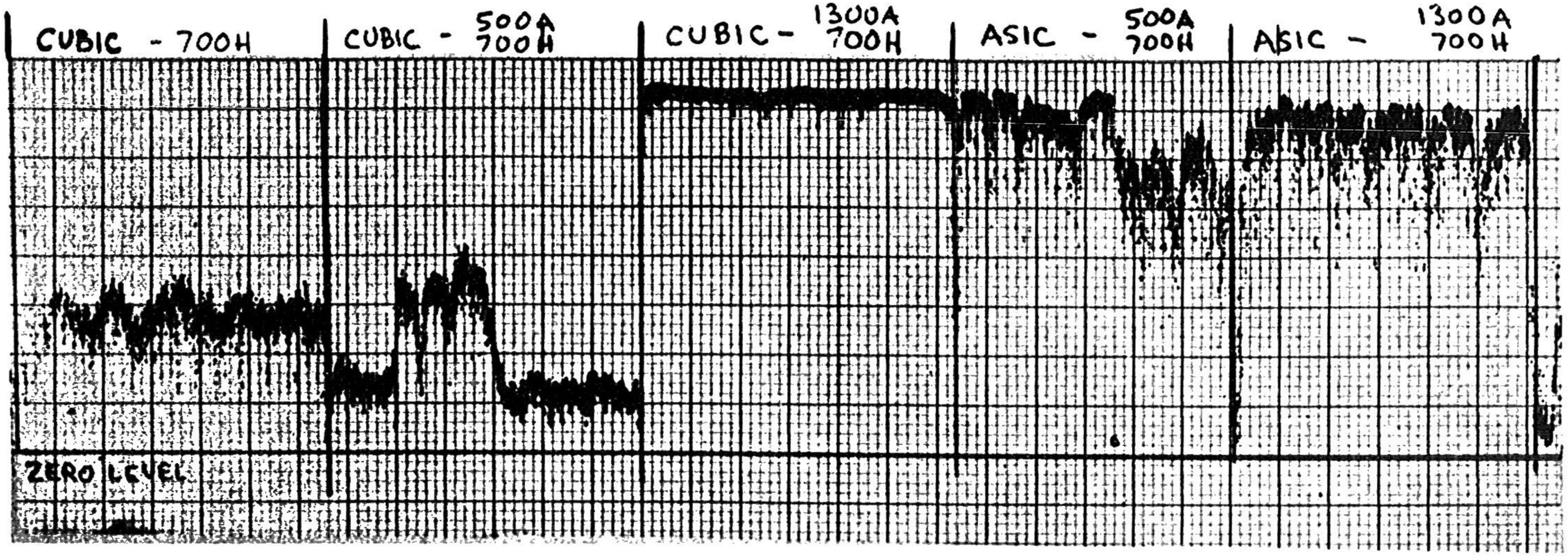


Plaintiff's Exhibit No. 2



RELATIVE OUTPUT OF 2000 CYCLE SIGNAL

Plaintiff's Exhibit No. 3



RELATIVE OUTPUT OF 4000 CYCLE SIGNAL

UNITED STATES COURT OF APPEALS FOR THE
DISTRICT OF COLUMBIA CIRCUIT

ARMOUR RESEARCH FOUNDATION OF)
ILLINOIS INSTITUTE OF TECHNOLOGY,)
Plaintiff-Appellant)
v.) Appeal No. 14, 550
ROBERT C. WATSON,)
Commissioner of Patents)
Defendant-Appellee)

STIPULATION AS TO CONTENTS OF JOINT APPENDIX TO BE PRINTED

MAY IT PLEASE THE COURT:-

It is hereby stipulated by and between the parties hereto, by their respective counsel, that the following parts of the record in Civil Action No. 2084-56, Armour Research Foundation of Illinois Institute of Technology v. Robert C. Watson, Commissioner of Patents, in the United States Court for the District of Columbia, be printed in a separate joint appendix:-

(1) The Complaint for Issuance of Patent filed in said District Court May 17, 1956.

(2) The Answer to the Complaint filed in said District Court July 3, 1956.

(3) The entire official Transcript of Proceedings in said District Court filed February 17, 1958.

(4) The Memorandum of Honorable Joseph C. McGarraghy, Judge of said District Court, dated March 31, 1958.

(5) Judgment of Honorable Joseph C. McGarraghy, Judge of said District Court, dated April 30, 1958.

(6) Findings of Fact and Conclusions of Law, dated April 30, 1958.

(7) Notice of Appeal to the United States Court of Appeals for the District of Columbia Circuit filed May 22, 1958.

(8) Designation of Contents of Record on Appeal, filed in said District Court June 4, 1958.

(9) Order to Transmit Original Exhibits from said District Court to said Court of Appeals, filed June 27, 1958.

(10) Copy of the specification of the Marvin Camras Patent Application Serial No. 114, 677 entitled "Improved Coating Material for Magnetic Recording", which specification is a part of Plaintiff's Exhibit 1 in said District Court, excluding all of the twenty (20) originally submitted and now cancelled claims, and including the following changes made in the originally filed specification by subsequently filed amendments, in order that said specification will appear in the Joint Appendix in the exact status that it was at the date of May 17, 1956 when the aforesaid Complaint was filed in said District Court:

(a) the change made in line 22 of page 3 by the amendment of June 22, 1951 (Amendment B, Paper No. 4).

(b) the changes made in the last line of page 4 by the amendment of December 4, 1950 (Amendment A, Paper No. 3).

(c) the changes made in lines 2 through 5 of page 5 by the amendment of December 4, 1950 (Amendment A, Paper No. 3) including the substitute sentence inserted thereby.

(d) the insert made in line 14 of page 6 after "like" by the amendment of April 29, 1954 (Amendment E^1, Paper No. 11).

(e) the changes made in line 4 of page 8 by the amendment of June 22, 1951 (Amendment

B, Paper No. 4).

(f) the changes made in the third line from the bottom of page 13 by the amendment of December 4, 1950 (Amendment A, Paper No. 3).

(11) Certified copy of claims 35 through 39 submitted in the amendment of November 1, 1954 (Amendment G^1, Paper No. 15) in the Marvin Camras Patent Application Serial No. 114, 677 entitled "Improved Coating Material for Magnetic Recording".

(12) The Brief on Appeal re Appeal to Board of Appeals, U.S. Patent Office, in said Camras application, which Brief was filed March 1, 1955.

(13) The Examiner's Answer re Appeal to Board of Appeals, U.S. Patent Office, in said Camras application, which Examiner's Answer is dated May 25, 1955.

(14) The Reply Brief re Appeal to Board of Appeals, U.S. Patent Office, in said Camras application, which Reply Brief was filed June 13, 1955.

(15) The decision of the Board of Appeals re Appeal to Board of Appeals, U.S. Patent Office, in said Camras application, which decision is dated March 8, 1956.

(16) The Petition for Rehearing and Reconsideration re Appeal to Board of Appeals, U.S. Patent Office, in said Camras application, which Petition was filed March 27, 1956.

(17) The decision of the Board of Appeals re Petition for Rehearing and Reconsideration filed March 27, 1956, which decision is dated April 18, 1956.

(18) The affidavit of the Marvin Camras filed as a part of Paper No. 15 aforesaid, in the said Camras application.

(19) References cited during the prosecution of Camras Patent Application Serial No. 114, 677 in the U. S. Patent Office, included in Defendant's Exhibit 1 in said District Court and comprising:-

(a) Camras U. S. Patent No. 2, 694, 656.

(b) Abraham et al article in "Nature" page 930.

(c) Mellor, "Treatise on Inorganic and Theoretical Chemistry", Volume 13, pages 732, 736, 738 and 780.

(d) Welo and Baudisch, "Two Stage Transformation of Magnetite into Hematite", pages 399 through 408.

(20) Chart of Relative Output at 2, 000 Cycles, Plaintiff's Exhibit 2 in said District Court.

(21) Chart of Relative Output at 4, 000 Cycles, Plaintiff's Exhibit 3 in said District Court.

(22) This Stipulation as to Contents of Joint Appendix to be Printed.

(23) Docket entries.

/s/ Benjamin H. Sherman

July 18, 1958

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July 18, 1958

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CIVIL DOCKET
UNITED STATES DISTRICT COURT FOR THE DISTRICT OF COLUMBIA

Parties	Attorneys	Number
Armour Research Foundation of Illinois Institute of Technology v	Smith, Michael & Gardiner, Woodward Bldg.	2084-56 ACTION FOR Letters Patent
Robert C. Watson Commissioner of Patents	C. W. Moore U. S. Patent Office	

Date	Account	Rec'd.	Disb'd.
1956			
May 17	Gardiner	$10.00	
May 17	U. S. Treas.		$10.00
1958			
May 22	Smith	5.00	
May 22	U. S. Treas.		5.00

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Date	Proceedings
1956	
May 17	Complaint, appearance filed
May 17	Summons, copies (1) and copies (1) of Complaint issued Ser. U. S. Atty. 5-17-56. Ser. Atty. Gen. 5-23-56.
July 3	Answer of deft. to complt. c/m 7-2-56. App. C. W. Moore filed.
July 3	Calendered (N)
1958	
Jan. 15	Hearing commenced, concluded and submitted. (Rep. M. A. Deeds) McGarraghy, J.
Feb. 4	Brief for deft. c/m 2-4-58 filed.
Feb. 4	Stipulation of counsel as to corrections in spelling in Official Transcript filed.
Feb. 4	Post-Trial Brief of pltff. ser/ack 2-4-58, exhibit I. filed.
Feb. 13	Reply brief of deft. c/m 2-13-58. filed
Feb. 13	Reply brief of pltff. c/s 2-13-58. filed
Feb. 17	Transcript of proceedings of Jan. 15, 1958 (Clerk's copy)filed
Feb. 21	Memorandum opinion dismissing complaint (Order to be presented.) McGarraghy, J.
Apr, 30	Findings of fact and conclusions of law (N) filed.
Apr. 30	Judgment dismissing complaint (N) McGarraghy, J.
May 15	Receipt for withdrawal of pltff's exhibit filed.
May 15	Trial brief of pltff. filed.
May 22	Notice of appeal of pltff. from order 4-30-58 (notice to C. W. Moore) deposit by Wm. A. Smith, Jr. $5.00 filed.
May 22	Cost Bond on appeal of pltff. in amount of $250.00 with USF&G approved and filed.
June 4	Designation of record on appeal by pltff. ser. ack. 6-4-58 filed.